Deliverable 6.4
Thermal Treatment of Solid Radioactive Organic Wasteforms

Date 31.07.2024 Version Final

Dissemination level: Public

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This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 945098.

Project acronym PREDIS	Project title PRE-DISposal management of radioactive waste	Grant agreement No. 945098
Deliverable No. D6.4	Deliverable title Thermal treatment of solid radioactive organic waste forms.	Version Final
Type Report	Dissemination level Public	Due date M47
Lead beneficiary Galson Sciences Ltd.		WP No. 6
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Abstract

This report summarises the types of Radioactive Solid Organic Waste (RSOW) requiring treatment and reviews and summarises the thermal treatment and conditioning technologies suitable for the treatment of RSOW that are available or under development. The scope of this review includes thermal treatment technologies that were studied in the course of the PREDIS project and other technologies which are available, or under development, globally.

The following treatment technologies studied within PREDIS have been reviewed and summarised:

- Gasification
- Hot Isostatic Pressing (HIP)
- Molten Salt Oxidation (MSO)
- Plasma processing
- Wet oxidation
- Pyrolysis

Additionally, the following technologies not studied within PREDIS have been reviewed and summarised:

- Acid digestion
- Supercritical water oxidation
- Thermochemical treatment
- Thermal desorption
- Vitrification

Keywords

Thermal Treatment, Value Assessment, RSOW

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Notification

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Acknowledgement

This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 945098.

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LIST OF ACRONYMS

AFIC	Active Furnace Isolation Chamber
ANSTO	Australian Nuclear Science and Technology Organisation
BWR	Boiling Water Reactor
CFB	Circulating Fluidised-Bed
CCIM	Cold Crucible Induction Melter
EDTA	Ethylenediaminetetraacetic acid
EC	European Commission
H&S	Health and Safety
HEPA	High-Efficiency Particulate Air
HIP	Hot Isostatic Pressing
HTO	Hydrothermal Oxidation
IHC	Induction, Hot Credible
ILW	Intermediate Level Waste
IAEA	International Atomic Energy Agency
IERs	Ion Exchange Resins
IPA	Isopropyl Alcohol
ISA	Isosaccharinic Acid
JHCM	Joule-Heated Ceramic Melter
KAERI	Korean Atomic Energy Research Institute
LLW	Low Level Waste
MDF	Materials Detritiation Facility
MSO	Molten Salt Oxidation
PPE	Personal Protective Equipment
PACT	Plasma Arc Centrifugal Treatment
PCBs	Polychlorinated Biphenyls
PTFE	Polytetrafluoroethylene
PVC	Polyvinyl Chloride
PMF	Powdered Metal Fuel
PHWR	Pressurised Heavy Water Reactor
PWR	Pressurised Water Reactor

PRIME	Pyrolysis Resins in Mobile Electric installation
RSOW	Radioactive Solid Organic Waste
RAG	Red Amber Green
SVOC	Semi Volatile Organic Compounds
SWORD	Springfields Waste Organic Residue Digester
SCWO	Supercritical Water Oxidation
TRL	Technology Readiness Level
THOR	Thermal Organic Reduction
US DOE	US Department of Energy
NSWC	US Naval Surface Warfare Centre
VTD	Vacuum Thermal Desorption
VES	Vinylesterstyrene
VOC	Volatile Organic Compounds
WAC	Waste Acceptance Criteria

1 Introduction

Radioactive Solid Organic Waste (RSOW) is a broad category of radioactive waste that comprises any waste containing a significant component of organic material. The term "RSOW" can be used to refer to a variety of conditioned and non-conditioned wastestreams produced by various waste producers and their physio-chemical characteristics may differ greatly. RSOW streams can present a number of management challenges, particularly where conventional conditioning methods produce wasteforms whose safe long-term storage and disposal is difficult to achieve and demonstrate, because they are considered not sufficiently stable in the conditions expected in a final repository.

Work Package six “Innovations in solid organic waste treatment and conditioning” (WP6) of the PREDIS project was concerned with the treatment and conditioning of RSOW. In WP6, options for the treatment and conditioning of thermally treated RSOW were developed and investigated.

The original deliverable title of D6.4 was “Implemented database: Matching the chemical composition of the reconditioned wastes” (Confidential for the consortium). However, early on in the project (early 2022) it was found in the Task 6.2 “Gap analysis and database on solid organic waste forms and their final state and value assessment analysis” that the work had to be rescoped, as the target originally specified expanding a database from the THERAMIN project. The database couldn’t be used as it was a partners-only deliverable. Therefore, the current deliverable is the outcome of the resulting rescope with the available data.

1.1 Aims and Objectives

The aim of this report was to summarise the types of RSOW requiring treatment and review and the thermal treatment and conditioning technologies suitable for the treatment of RSOW that are available or under development.

The scope of this review includes:

- Solid organic Low Level Waste (LLW) and Intermediate Level Waste (ILW).
- Thermal treatment technologies that were studied in the course of the PREDIS project and other technologies which are available, or under development, globally.

2 Sources of RSOW

The first part of this task considered the types of RSOW which may benefit from thermal treatment. This section describes the main categories of RSOW that could be envisioned to benefit from novel treatment approaches, and the disposability risks associated with each waste type.

2.1 Ion Exchange Resins

Ion Exchange Resins (IERs) are used in a liquid purification role in many processes in the nuclear industry; details on the application of ion exchange processes to radioactive waste and the treatment of ion exchange materials can be found in an extensive review on the subject undertaken by the IAEA[1]. Ion exchangers function by exchanging ions bound into the solid matrix of the resin with mobile ions in an external liquid solution. There are several different types of ion exchange media including inorganic ion exchangers (such as clays and zeolites) and naturally occurring organic ion exchangers (such as charcoal), but the most used type, and the type which is of interest in this report, are synthetic organic ion exchange resins. These resins typically comprise a cross-linked matrix of hydrocarbon chains (polystyrene divinylbenzene is the most common matrix) in which different functional groups may be embedded. Contaminant ions in solution are removed when they exchange with ions associated with accessible functional groups in the matrix.

IERs can be classified by whether they provide exchange sites for negatively charged ions (anionic resins) or positively charged ions (cationic resins). The functional group will define the acidity (for cation exchangers) or alkalinity (for anion exchangers) of the resin. IER beds may consist of only one type of resin or may be operated in a ‘mixed bed’ system in which both anionic and cationic resins are mixed in order to remove both positively and negatively charged ions from solution.

Organic ion exchange resins are used in a number of liquid purification processes, the most common nuclear applications are:

- Purification of coolant (both primary and secondary loops) for water-cooled reactors,
- Treatment of fuel storage pond water, and;
- Treatment of radioactive aqueous liquid effluent.

Due to the generation of spent organic ion exchange resin by routine reactor operations, all states that operate water cooled power reactors generate some volume of spent IERs. Typical generation rates for spent organic IERs by different types of reactors are shown in Table 2.1.

Table 2.1:Typical spent organic ion exchange resin generation rate by reactor type[1].

Reactor Type	**Spent ion exchange resin generation ($m^3 \cdot unit^{-1} \cdot a^{-1}$)**
Pressurised Water Reactor (PWR)	4-7
Pressurised Heavy Water Reactor (PHWR)	5-7
Boiling Water Reactor (BWR)	20

Organic IERs present a number of storage and disposability issues in the form that they arise. In particular:

- IERs arise wet and require a dewatering or drying step to be suitable for further conditioning or treatment.
- Spent IERs may have very high activities, necessitating shielding during transport and operations. IER activities also may not meet the Waste Acceptance Criteria (WAC) of disposal facilities (resins with significant alpha activity are often particularly problematic for surface and near-surface facilities).

- Radiolytic and biological degradation of IERs can produce mobile or acidic products which may not be compatible with the WAC of disposal facilities.
- Radiolytic and biological degradation of IERs can produce gases such as hydrogen and methane. The production of gases can present a number of hazards during storage, transport and disposal, including pressurisation and fire/explosion if not ventilated sufficiently.
- IERs can swell in contact with water, which can compromise the stability of the conditioning matrix.
- Spent IERs can contain chemical components that make them incompatible with cementation or otherwise unsuitable for disposal. Examples include:
    - IERs that contain boric acid (potentially arising from PWR primary circuit purification), which adversely affects the setting of cement.
    - IERs containing Ethylenediaminetetraacetic acid (EDTA), which acts as a complexant for radionuclides and may not be compatible with the WAC of disposal facilities.

Additionally, spent IERs often do not meet the WAC for incinerators due to, for example, the presence of significant alpha-emitting or volatile radionuclides. As a result of the disposability issues and the potential incompatibilities with conventional incinerators, IERs can benefit significantly from alternative thermal treatment technologies.

2.2 Mixed Waste

The largest volumes of solid organic waste typically arise in the form of lightly contaminated waste produced as a byproduct of the general operations and maintenance of power plants and research centres[2]. This waste may be comprised of various forms of general waste including used Personal Protective Equipment (PPE), wipes, sheeting, lab equipment, containers, cable insultation and tools[3]. The solid organic component of this waste typically consists of a mixture of various kinds of synthetic polymer (including plastics and resins), rubber, paper and cotton products. The organic component of the waste is often mixed with non-organic components such as metal, glass and ceramic.

Some of the primary storage and disposability issues related to mixed solid organic waste are:

- Fire hazard, arising from the combustible nature of typical organic wastes.
- Gas production, from the degradation of organic materials by radiolytic and biological processes.
- Production of liquids by radiolytic and biological degradation processes. The liquid phase can present a potential transport pathway for radionuclides.
- The creation of voidage in the disposal facility due to the reduction in volume from the degradation of organic materials.
- The presence of halogenated polymers (such as polyvinyl chloride (PVC), polytetrafluoroethylene (PTFE) and neoprene). PVC is typically the largest single component of mixed organic radioactive waste[3], with common PVC products including protective suits, bagging material and sheeting. The decomposition of chlorinated plastics such as PVC under disposal conditions may produce hydrochloric acid, which can chemically attack componants of the facility and locally compromise the pH buffering function of the backfill in certain repository concepts.
- The presence of cellulose, which is present in radioactive waste containing wood or cotton products such as paper or cloth wipes, filters, and overalls. Under alkaline conditions (as may be generated by a cement encapsulant or backfill), cellulose can undergo hydrolysis, producing isosaccharinic acid (ISA) which can act as a complexant for a number of radionuclides, increasing their mobility[3]. Consequently, a number of repositories have limits on the mass of cellulose containing materials that can be disposed of specified in their WAC.

Mixed organic waste is generally compatible with conventional thermal treatment (incineration), although some waste streams may have features which preclude this route; features of mixed organic waste that may be incompatible with conventional incineration include:

- The presence of significant quantities of alpha-emitting radionuclides.
- The presence of radionuclides which will evolve into a gas during incineration (including tritium, ^{14}C, and volatile radionuclides).
- The presence of large volumes of non-combustible waste in the waste stream (metals, concrete, glass).
- The presence of components which may evolve acidic gases under incineration (particularly PVC). The evolution of acids can increase off-gas handling requirements or reduce the lifetime of components of the incinerator.

2.3 Conditioned Waste

Conditioned waste has been solidified into a stable wasteform, typically with the intent to provide a package suitable for storage and disposal. Types of conditioned waste with a solid organic component are presented in the sections below. Conditioned wasteforms are usually designed to meet storage and disposal requirements and consequently generally do not require further treatment. There are, however, a number of situations in which an already conditioned package may be considered for additional treatment; reasons why conditioned waste may require further treatment include:

- Packages conditioned for historical disposal practices (e.g. sea disposal) or historical WAC that are unsuitable for modern facilities.
- Packages that have degraded due to age or storage conditions.
- Packages that do not meet facility WAC due to errors in the management process such as mischaracterisation of the chemical or radiological content of the waste or incorrect formulation of the wasteform matrix.

The primary classes of conditioned waste that may be considered for thermal treatment are described in the following sections.

2.3.1 Bitumen encapsulated waste

Bitumen is an organic material that is used as an encapsulant in a number of countries[3,4]. Commercially available bitumen is produced as the bottom product following the distillation of crude oil. Bitumen encapsulation is undertaken by mixing waste with a heated bitumen encapsulant and allowing it to cool, resulting in a solid, hardened product suitable for transport and disposal. Wastes suitable for bitumen encapsulation include soluble salts, ion exchange resins, sludges and incineration ashes.

2.3.2 Polymer encapsulated waste

Polymer encapsulation entails encapsulating a waste in a polymer. The adoption of polymer encapsulation is generally motivated by properties of the waste that would make it incompatible with cement encapsulation. In particular polymers typically provide an effective barrier to water and so are useful for encapsulating waste with components that will react with or dissolve in water. Polymers are used to encapsulate spent IERs[1], reactive metals (such as Al, Mg and U) and water-soluble residues.

The most commonly used polymers for radioactive waste encapsulation are vinylesterstyrene (VES) and various epoxy resins[3]. The polymer encapsulation process typically involves mixing a number of polymer-forming components together, which undergo a polymerisation reaction and harden over a period of a few minutes to a few days. An alternative process used for thermoplastic polymer encapsulants (such as polyethylene) involves melting the polymer to achieve a fluid state suitable for mixing with waste.

2.3.3 Cement encapsulated organic waste

Cement encapsulation is one of the most common conditioning methods for low and intermediate level waste[2]. Broadly there are two approaches to cement encapsulation adopted for the conditioning of organic waste:

- Direct cementation, where the waste is cemented into a container. Depending on the nature of the waste and process considerations this may involve mechanical mixing of the cement and waste component to achieve an intimate mixture or simply ‘flood grouting’ and relying on the fluid properties of the cement to infiltrate the waste. Direct cementation is sometimes used for the conditioning of ion exchange resins.
- Compaction and cementation, whereby the waste is initially compacted before being loaded into a package and cemented. Typically, a sacrificial drum is used for the compaction step and a number of the resultant compacted ‘pucks’ are loaded into the disposal drum and grout poured around them. Compaction and cementation are often used for mixed organic wastes as they typically have a high compaction factor.

Cement encapsulated organic waste is generally incompatible with conventional incineration due to the presence of significant quantities of non-combustible cement.

3 Thermal Treatment and Conditioning Technologies

The thermal treatment or conditioning of RSOW can realise a number of benefits, including significant volume reduction and remediating a number of the disposability and storage issues present for untreated RSOW, as identified in Section 2. Conventional incineration, consisting of the combustion of the waste in air, is already widely used for the treatment of RSOW, although there are wastestreams for which it is not appropriate. This section describes alternative thermal technologies for RSOW which may be of benefit to national waste management programmes.

The TRL of each technology is evaluated according to the publicly available information found during this review according the TRL definitions set out in the General Annexes of the HORIZON 2020 work programme[5]. The following definitions apply:

- **TRL 1** – basic principles observed.
- **TRL 2** – technology concept formulated.
- **TRL 3** – experimental proof of concept.
- **TRL 4** – technology validated in lab.
- **TRL 5** – technology validated in relevant environment (industrially relevant environment in the case of key enabling technologies).
- **TRL 6** – technology demonstrated in relevant environment (industrially relevant environment in the case of key enabling technologies).
- **TRL 7** – system prototype demonstration in operational environment.
- **TRL 8** – system complete and qualified.
- **TRL 9** – actual system proven in operational environment (competitive manufacturing in the case of key enabling technologies).

3.1 Thermal Technologies Studied in PREDIS

Within WP6, six thermal treatment routes are considered, which result in either the production of glassy or ceramic materials without the need for additional treatment, or in the generation of treated wastes that need to be immobilised using matrices made of cement or geopolymer. These technologies are described in the following datasheets:

- Gasification
- Hot Isostatic Pressing (HIP)
- Molten Salt Oxidation (MSO)
- Plasma processing
- Wet oxidation
- Pyrolysis

3.1.1 Gasification

Title: Gasification

Type of process: Thermal treatment

Description of process:
The gasification process involves heating waste in a low oxygen environment where it undergoes partial oxidation and combustion[6,7]. The process converts the organics in the waste into a synthetic hydrocarbon gas (syngas), composed primarily of CO, H and CH_4. The produced gas contains significantly less unwanted SO_x and NO_x than is produced by incineration. Any non-combustible components are left as a glassy ash residue with low organic content. The syngas can potentially be burned for heating, electricity generation, or for powering the treatment plant.

The gasification process was first developed for producing coal gas but has since been modified to process radioactive waste. VTT has patented a gasification-based thermal treatment process to treat low and intermediate level radioactive wastes containing organic matter[8,9]; the principle of the process is illustrated in Figure 1, and depiction of the gasification rig is given in Figure 2.

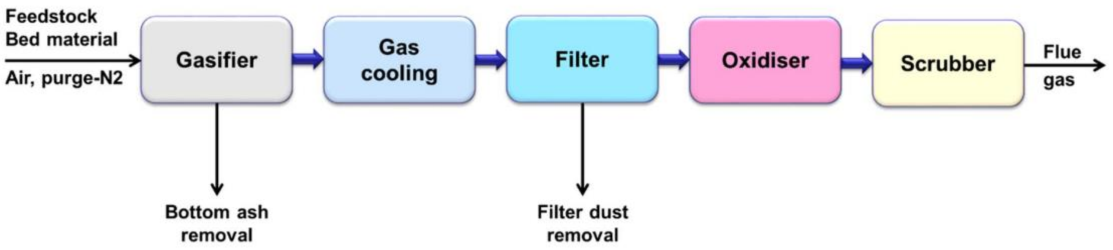

Figure 1. Flow diagram for the gasification-based thermal treatment facility patented by VTT[9].

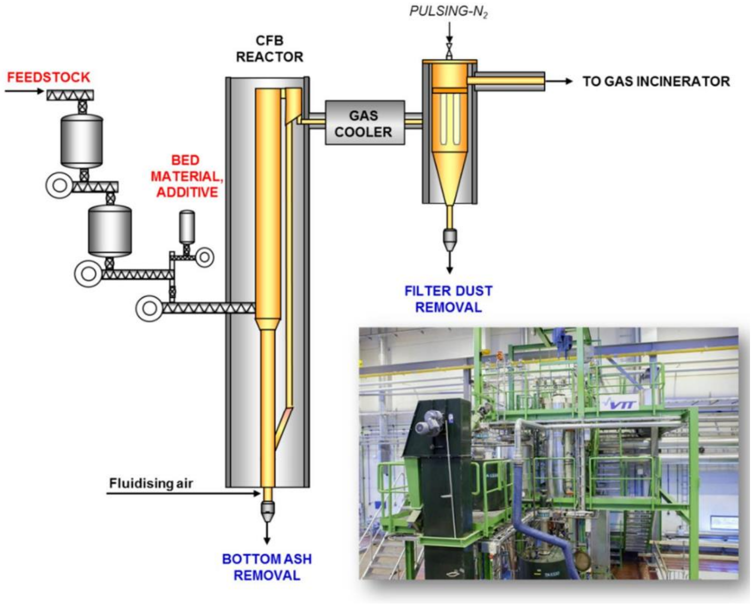

Figure 2. VTT's pilot-scale Circulating Fluidised-Bed (CFB) gasification rig[9].

Resource requirements:

- Energy for heating (comparable to incineration).

Value assessment considerations

- Health and Safety (H&S): Risks due to high temperature and activity concentration. Risk of liberating gaseous radioactive components.
- Risk and Hazard reduction: Organics in the waste are destroyed and converted into an ash.
- Environment: Significant energy cost, partially offset by the production of a syngas which (once scrubbed of contaminants) can be used for heating, electricity generation, or for powering the treatment plant.
- Socio-economic: Volume of waste reduced, lowering storage and disposal costs.

Type of waste material that can be processed (input):

Suitable RSOW waste groups:

- Spent IERs.
- Mixed organic waste.
- Waste conditioned in an organic matrix (bituminised and polymerised waste) may be suitible.

Other suitable wastes:

- Oily waste.
- Sludge.
- Uranium contaminated waste[10].

Constraints and Exclusions:

Non-combustible materials, such as metal, cement and glass are unsuitable as they can be detrimental to the performance of the system (reducing the temperature). Unsuitable for wastes containing significant quantities of volatile radionuclides.

Type of material produced by the process (output):

A syngas is produced which can be used for generating thermal energy or electricity.

A glassy ash-like material containing a low amount of carbon is left as a residue, which contains the non-volatile radionuclides from the waste. The ash will require treatment to be made suitable for disposal.

Secondary waste:

Off-gas treatment expected to be required, generating associated secondary wastes.

Is the product likely to need further conditioning prior to disposal?

Ash product requires further treatment, e.g. immobilisation in geopolymers.

Current international practices:

Gasification has not been used for radioactive waste treatment. A number of plants exist for treating municipal wastes. Small mobile systems are commonly employed on ships and for disposing of hazardous medical or military wastes.

VTT has proposed to use their gasification plant[8,10] for treating low level wastes but the system is still at the pilot scale. Inactive trials have been completed within the European Commission (EC) THERAMIN and PREDIS projects. Their results have shown very efficient removal of organic matter from ion exchange resins and significant volume reduction factors.

Facilities:

- Various conventional (inactive) systems.

• Gasification plant operated by VTT (Finland)[11].
Technical maturity: TRL 6 – technology demonstrated in industrial environment, tests have been undertaken on simulant waste at scale, although no tests on active waste at scale have been undertaken.
Work within PREDIS: Within the PREDIS project, SIIEG deployed a coupled plasma-gasification process to thermally treat a surrogate spent cationic IER doped with Cs[12]. Development work focused on the optimisation of the reaction temperature to ensure conversion of the mobile and volatile Cs species into inorganic and thermally stable compounds. VTT Finland have used thermal gasification to treat a mixed bed IER to produce ash. The tests were done using an atmospheric pressure bench-scale bubbling fluidised bed gasifier. The IER was doped with Cs, Eu, Ce, and Fe tracers. The resulting ashes had a factor of 14 concentration of Ce relative to the waste feed, a concentration factor of 33 for Cs and a factor of 7 for Eu. This indicated that certain types of activity are held up in the process vessel.

3.1.2 Hot Isostatic Pressing

Title: Hot pressing / Hot Isostatic Pressing

Type of process: Conditioning

Description of process:
Hot Isostatic Pressing (HIP) consists of applying a uniform high pressure to a waste at high temperature in order to immobilise a waste into a compacted can of glass-ceramic or ceramic wasteform. A typical process flow for HIP is outlined below[13]:

1. The feedstock is calcined to remove moisture.
2. The calcine is milled along with a precursor or glass frit.
3. The resulting fine, granualar material is packed into a thin-walled stainless steel vessel (a `HIP can’).
4. The HIP can is then evacuated and sealed.
5. The can is HIPed; it is placed in a HIP vessel and an inert gas blanket (such as argon) is used to uniformly apply heat and pressure to the sealed can. Pressures and temperatures on the order of 100 MPa and 1000°C result in the compaction of the can and contents and the production of a densified glass-ceramic or ceramic wasteform[14].

Resource requirements
- Energy for heating (more than incineration).
- Inert gas (re-usable).
- Precursor/frit (dependant on waste and approach).

Value assessment considerations
- H&S: Risks due to high temperature, high pressure and activity concentration. Batch process provides good criticality control. HIP can provides containment of radionuclides, including volatiles.
- Risk and Hazard reduction: The volume of the waste is reduced, good containment is provided by the HIP can and wasteform.
- Environment: Significant energy cost, compounded by the need for multiple thermal treatment steps.
- Socio-economic: The volume of the waste is reduced, lowering storage and disposal costs. Provides a potentially a cheaper alternative to vitrification[15].

Type of waste material that can be processed (input):
Suitable RSOW waste groups:
RSOW is not suitable for HIPing in its raw form and requires an initial thermal treatment step prior to HIPing. HIPing has been demonstrated for the treatment of ashes from the pyrolysis of spent IERs and mixed organic waste.

Other suitable wastes:
- Bead/powder inorganic ion exchangers.
- Pond sludges (trials conducted for magnox sludge[16]).
- Plutonium and plutonium residues[17].
- Reprocessing calcines[15].
- Uranium-containing wastes[13,18].

Constraints and Exclusions:
The process cannot directly treat organic wastes (these must be calcined or otherwise thermally treated before HIPing) and only a small amount of metal can be accepted. The maximum size of the HIP can is constrained by the requirement for uniform pressing of the product.

Type of material produced by the process (output):
The final product is a dense glass-ceramic or ceramic wasteform in a steel HIP can.

Secondary waste:
Filters from the off-gas system (pre-treatment).

Is the product likely to need further conditioning prior to disposal?
Yes, the HIP cans would typically require some form of consolidation into a larger package for storage and disposal due to the lack of handling features and deformation of the can following HIPing.

Current international practices:
Small-scale active and inactive test facilities in the UK use the technique for the immobilisation of a diverse range of wastes arising from nuclear fuel processing activities on the Sellafield site. Small-scale plant for the immobilisation of Pu residues has been taken to preliminary design and considered TRL 5-6 for those waste feeds.

Australian Nuclear Science and Technology Organisation (ANSTO) is undertaking commissioning for a plant to treat radioactive wastes arising from Mo isotope production, an active facility is due to begin operations in 2025[13].

The University of Sheffield (UK) have the capacity to use a conventional HIP set up with an Active Furnace Isolation Chamber (AFIC) to treat radioactive wasteforms on a prototype scale[19].

The US Department of Energy (US DOE) has experimented processing a non-radioactive simulated plutonium wasteform using this technique. The trials were successful in part and with further modifications could be potential final solution for plutonium immobilisation[20].

Facilities:

- Industrial scale active facility in comissioning at ANSTO (Australia).
- Various conventional (inactive) systems.

Technical maturity:
TRL 7 – Industrial scale plant for commercial radioactive waste treatment is being commissioned but is yet to start operations (ANSTO). TRL 5-6 within Europe.

Work within PREDIS:
Within PREDIS, the University of Sheffield has undertaken trials into the HIPing of RSOW and products of RSOW treatment[21]. The following cases were tested:

- The HIPping of untreated IERs and polyethylene. HIPping of the untreated RSOW was impossible as the welding of the can resulted in the thermal decomposition of the polymers, resulting in the expulsion of waste from the unfinished weld.
- The HIPping of sludge from the wet oxidation of an IER simulant (see wet oxidation datasheet). Sludges comprising different ratios of Fe_2O_3 and CuO were

dried and thermally treated prior to HIP. The HIP was successful, although the products require further characterisation.

- The HIPping of ashes produced by the pyrolysis of simulant mixed orangic waste and IERs by the IRIS process (see pyrolysis datasheet). Four approaches to HIPping of these ashes were tested:
  - Direct HIPing of ashes as received.
  - HIPing of the ground ashes.
  - HIPing of the ground ashes with sodium tetraborate ($Na_2B_4O_7$).
  - HIPing of the ground ashes with sodium aluminate ($NaAlO_2$).

  All of the trail cases formed solid, densified products, although the directly HIPed ashes exhibited relatively high porosity and only the sodium tetraborate case formed a true glass-crystalline product.

Trials were also undertaken on the HIPing of the same ashes at NNL, although these were unsuccessful at the larger scale due to difficulty in fully drying the ashes following a wet milling step. Water was used to create a suspension of ashes for wet milling and following milling the ashes were not sufficiently dried prior to being HIPed resulting in a product which was not properly densified.

Additionally, KIPT investigated the HIPing of contaminated wood ash. Research at KIPT was interrupted due to the invasion of Ukraine, and the prepared canisters were instead sent to the University of Sheffield. The HIP of the ashes was ultimately unsuccessful due to a suspected pinhole leak in the can welds.

3.1.3 Molten Salt Oxidation

Title: Molten carbonate treatment / Molten salt oxidation

Type of process: Treatment

Description of process:
Molten Salt Oxidation (MSO) entails oxidising waste within a vat of molten carbonate salts at temperatures between 400 and 900°C[1,22,23]. Air or oxygen is pumped into the salt bath and oxygen reacts with the carbonate to produce free peroxide and superoxide ions which then react with the waste. The organics are oxidised, with any metals or inorganic residue retained in the salt melt. An advantage of MSO over conventional incineration is that acidic gases, produced for example by the decomposition of halogenated organics, react with the carbonate melt and are retained as a salt (this property also allows MSO to be used as part of an off-gas management system as an alternative to wet scrubbing[23]). Typically, only steam, oxygen, nitrogen and CO_2 (which can include C-14) are produced as off gases in significant quantities (as they do not react with the melt), significantly reducing the burden for filtering and scrubbing compared to incineration or pyrolysis. This process results in virtually complete removal of all the organics. The residue left behind by the waste builds up in the salt, reducing its fluidity and ability to absorb acidic gases. This necessitates the discharge of salts once they are ‘spent’ (typically at ash contents of about 20 wt%). When the salts are cooled, the inorganic components, including radionuclides, are bound within the crystallised salts.

Resource requirements:
- Carbonate salts (usually Na_2CO_3, K_2CO_3, Li_2CO_3, or a mixture of these salts).
- Energy for heating (less than incineration).

Value assessment considerations
- H&S: Risks due to high temperature and activity concentration.
- Risk and Hazard reduction: Organics in the waste are destroyed, although spent salts pose some disposability challenges as they are not compatible with conventional cements.
- Environment: Relatively lower energy cost as compared to incineration. Acidic gases are retained in the salt bed.
- Socio-economic: Volume reduction resulting in lower storage and disposal costs.

Type of waste material that can be processed (input)[24,25,26]:
Suitable RSOW waste groups:
- Mixed organic waste, including halogenated plastics and cellulosic waste.
- IERs.

Other wastes:
- Chlorinated solvents.
- Oils.
- Biological waste.
- Cellulosic waste.
- Plutonium contaminated waste.
- Uranics, including spent fuel.

Constraints and Exclusions:
Wastes where C-14 or tritium are major radionuclides.

Type of material produced by the process (output):
Spent salt containing inorganic residue of the waste.

Secondary waste:
The off-gas is relatively clean, so only small amounts of secondary waste from the off-gas system are expected.

Is the product likely to need further conditioning prior to disposal?
Yes – The salt will require further conditioning prior to disposal.

Current international practices

- Trials of molten salt oxidation were conducted in the USA at the Lawrence Livermore and Oak Ridge national labs. These trials successfully treated a range of organic wastes including low level radioactive materials and IERs[27].
- MSO is in use at the US Naval Surface Warfare Centre (NSWC) Indian Head Division for destruction of a wide range of inactive wastes. They have also conducted trials using the facility to treat Pu-contaminated analogues.
- CV Řež in Czechia operates an experimental MSO reactor with a focus on spent ion exchange resins and scintillator oils.
- The Korean Atomic Energy Research Institute (KAERI) has conducted bench scale inactive experiments using MSO to treat ion exchange resins[24].
- An MSO plant operates at the South Korean demilitarisation facility (DEFAC) to treat secondary wastes from munitions decommissioning. These include explosives, resins, and oils[28].

Facilities:

- Industrial scale plant for inactive waste at DEFAC (S. Korea).
- Industrial scale plant for inactive waste at NSWC Indian Head (USA).

Technical maturity:
TRL 6 – technology demonstrated in industrial environment and small-scale tests have been undertaken on radioactive waste.

Work within PREDIS:
Within PREDIS, the MSO process was used at CV Řež to treat spent IER surrogate. A two-stage MSO process was used where IERs are fed into a sodium carbonate salt bed in one vessel and the off-gas is routed through a second sodium carbonate salt bed in another vessel which serves to fully oxidise flue gases and capture fly ash.

3.1.4 Plasma processing

Title: Plasma processing

Type of process: Treatment / conditioning

Description of process:
Plasma processing utilises a plasma torch to heat and decompose waste materials[2]. Organic constituents are volatilised, pyrolysed, or combusted, while inorganic material and non-volatilised metals are melted into a pool. Volume reduction factors range from 6 (typical for the ZWILAG facility, Switzerland) for waste containing mostly metals and debris to 10 for treatment of mixed waste (typical values achieved at the Russian Pluton facility) and to more than 100 for primarily organic waste[29]. Existing plants typically treat solid organic wastes, but the technique could also be used for metallic and concrete containing waste[30].

The plasma is generated by the application of high voltage across a gas to ionise it and thus convert it into a plasma. Process conditions (in particular, the extent to which the treatment is oxidising) can be tuned by selection of the plasma gas. There are two main plasma torch types[31]:

- Transferred torches, where a plasma arc is created by a high potential difference maintained between the melt pool and an anode held just above the surface.
- Non-transferred torches, where a stream of working gas is ionised into a plasma inside a specialised chamber and injected into the working chamber in a plasma 'jet'.

The plasma torch may be coupled with other heating technologies to maintain a molten melt pool. Glass frit may be added into the crucible during treatment to yield a vitrified wasteform with enhanced properties (see vitrification data sheet). The gaseous products from the process are passed through an afterburner or catalytic converter for complete oxidation, followed by appropriate treatment of the off-gas.

Resource requirements:

- Plasma working gas.
- Electricity.

Value assessment considerations

- H&S: Risk due to high temperature. Specific activity of slag will be higher than original waste. Waste can be fed nearly unsorted, reducing exposure risks.
- Risk and hazard reduction: Destruction of organics and conversion into waste slag or vitrified product which is expected to be stable under storage and disposal conditions.
- Environment: High energy usage relative to incineration.
- Socio-economic: Relatively high capital and operating costs. Versatile, with a large range of potential feeds and good volume reduction, reducing storage and disposal costs.

Type of waste material that can be processed (input):
Suitable RSOW waste groups:

- Mixed organic waste, including halogenated plastics and cellulosic waste.
- IERs.
- Conditioned waste, including bituminised waste, cemented waste and polymerised waste.

Other suitable wastes:

- Oils.
- Sludges.
- Metals.
- Filters.
- Uranics.

Constraints and Exclusions:

Unsuitable for wastes containing significant quantities of volatile radionuclides.

Type of material produced by the process (output):

The end product is a solid slag or glass (if glass-forming materials are included). The product may be heterogeneous with different layers of different densities, although a homogeneous glassy product is achievable for certain waste feeds and process designs. If there are metals in the waste feed, a distinct metal layer is typically present in the product.

Secondary waste:

Secondary waste includes High-Efficiency Particulate Air (HEPA) filters, slag/sludges or aqueous solutions, all of which may require subsequent treatment. The process typically creates around 2% secondary waste by mass[30].

Is the product likely to need further conditioning prior to disposal?

No - the product is typically cast within metal or ceramic containers and suitable for direct disposal without any further treatment.

Current international practices:

Plasma melting is used routinely for commercial treatment of radioactive waste.

Facilities

- Plasma Arc Centrifugal Treatment (PACT) process (ZWILAG, Switzerland and Tsugura NPP, Japan)[32,33].
- Pluton plant (FSUE Radon, Russia)[32,34].
- Europlasma (Kozloduy NPP, Bulgaria)[35].
- PIVIC (CEA Marcoule, France) [prototype scale][36].
- SHIVA (CEA Marcoule, France) [prototype scale][36].

Technical maturity:

TRL 9 – Proven technology deployed for industrial scale treatment of radioactive waste.

Work within PREDIS:

Within PREDIS, the University of Sheffield has characterised the wasteform produced by the plasma vitrification of simulant plutonium contaminated waste drums[12]. The simulant waste consisted of 100 L drums containing different contaminated materials, including wood and other organics, masonry and construction materials, and PVC from packaging and handling. The drums were vitrified with added calcium aluminosilicate as a glass former to produce a stable, durable wasteform.

3.1.5 Wet oxidation

Title: Wet oxidation

Type of process: Treatment

Description of process:
Wet oxidation processes use a flameless chemical oxidation process to destroy organics in an aqueous solution. A typical wet oxidation process consists of mixing organic waste, hydrogen peroxide (H_2O_2), water and an inorganic catalyst at 100°C[37,38]. The organic fraction is oxidised to CO_2 which is released to an off-gas system. The inorganic contaminants (including the radionuclides) remain in solution. The aqueous effluent can be treated either by an existing aqueous treatment route or via evaporation to leave a concentrated inorganic solid.

A modified form of the technique uses sulphuric acid (H_2SO_4) as the oxidising agent rather than hydrogen peroxide[39].

Resource requirements:

- Chemical oxidant, H_2SO_4 or H_2O_2.
- Energy for heating (significantly less than incineration).

Value assessment considerations

- H&S: Risk due to the use of hazardous chemical oxidants. The process must be engineered to manage the heat generated by the chemical reaction.
- Risk & Hazard reduction: Destruction of organics.
- Environment: Relatively lower energy cost as compared to incineration. Has a relatively 'clean' gaseous exhaust.
- Socio-economic: Volume reduction (assuming drying of effluent), resulting in lower transport and disposal costs. Low capital costs.

Type of waste material that can be processed (input)[39]:
Suitable RSOW waste groups:

- Spent IERs.
- Mixed organics, including cellulosic waste.

Other wastes:

- Oily waste.
- Oily sludge.
- Uranium contaminated waste.

Constraints and Exclusions:
Solid waste must be in the form of a small particulate; large solid waste items are not suitable for wet oxidation.

Type of material produced by the process (output):
Inorganic residue which contains most of the radioactivity. This residue will require some form of immobilisation prior to disposal.

Secondary waste:
Off gas; possibly containing volatile radionuclides. Gas treatment plant may be required, and any scrubbers or filters required to treat the gaseous discharges will be secondary waste. Expect lower volumes of gas to require treatment than incineration and correspondingly lower volumes of secondary waste.

Is the product likely to need further conditioning prior to disposal?
Yes – product will require conditioning prior to disposal.

Current international practices:
Wet oxidation has been explored by a number of countries including Sweden, Italy, France, UK, USA and Japan.

Wet oxidation trials were undertaken by Sweden (by ASEA-ATOM) and Italy (by ENEA) in the 1980s and in France (by CEA) in the 1990s[40,41]. These trails successfully treated small volumes of ion exchange resins but did not progress to a large scale.

The ‘SWORD’ process has been used at the NNL Preston lab in the UK to treat uranium contaminated polishes and resins[39]. The process has also been adapted to treat tritiated oils (known as the TOAST process).

The company AEA Technology have developed and commercialised a wet oxidation process under the brand name ModulOx™[40,42]. A ModulOx plant has been built in the UK for commercial treatment of some small volume IER waste streams, this plant is due to begin active operations in 2024[43].

Facilities:

- Springfields Waste Organic Residue Digester (SWORD) (NNL Preston, UK).
- ModulOx process (UK).

Technical maturity:
TRL 7 – Industrial scale plant for commercial radioactive waste treatment (ModulOx) has been deployed but is yet to start operations.

Work within PREDIS:
Within PREDIS, surrogate IERs were treated using Fenton-Like wet oxidation by Polimi. Surrogate spent anionic and cationic resins were prepared by doping nuclear grade resins with Cl and I tracers (anion-exchange resin) and Co, Ni, Sr and Cs tracers (cation-exchange resins)[12]. The wet oxidation process used an iron-containing catalyst and a H_2O_2 oxidant. The precipitate and residue were mainly composed of inorganic sulphate compounds.

Wet oxidation was also studied by The University of Sheffield, a small bench-scale setup was used to treat fresh and simulant spent IERs. The resulting sludge was subsequently dried and densified by HIPping. An attempt was made to densify a simulant wet oxidation sludge at a larger scale at NNL, although this failed.

3.1.6 Pyrolysis

Title: Pyrolysis/gasification

Type of process: Treatment

Description of process:
Pyrolysis is a flameless dry distillation process which uses high temperatures to break down the organic components of wastes into new gases, liquids and solids (pyrolysates) under oxygen-free or oxygen-deficient conditions[44]. The organic fraction is not combusted (oxidised), instead complex organics are fractured into smaller components. The main outputs of pyrolysis are low mass hydrocarbons (methane, ethane, benzene and toluene), tars, and a carbon-rich char[31]. Pyrolysis has the advantage of having lower operating temperatures compared to incineration which allows it to retain relatively volatile radionuclides in the solid residue.

The pyrolysis processes can be operated as batch or continuous processes inside any process chamber which is able to be purged or evacuated of oxygen prior to heating. Common pyrolysis vessel designs include rotary kilns, fluidised beds and ‘pot’ type vessels. In-container treatment is also possible.

CEA’s IRIS process provides an example of a pyrolysis process designed for radioactive waste treatment[45]. IRIS is a three-step process implemented in rotating kilns where the first step consists of pyrolysis at 550°C, producing pitch that is then processed in a calcining step at 900°C in an oxygen-enriched atmosphere. The off-gases arising from the thermal treatments include a volatile hydrocarbon fraction that is oxidized at 1100°C in an afterburner. Before discharge to atmosphere, the gas stream is submitted to caustic scrubbing to eliminate the volatile acids. The initial pyrolysis step eliminates chlorine while at a low temperature, which limits corrosion problems. The subsequent calcination of the pitch results in a large volume reduction factor and the elimination of the remaining organics.

Pyrolysis is functionally very similar to gasification and calcination. Gasification uses the same process as pyrolysis (exposing organic material to high temperature under low oxygen conditions) but utilises higher temperatures optimised to produce syngas. The term calcination usually refers to the thermal treatment of non-organic materials under oxygen deficient conditions.

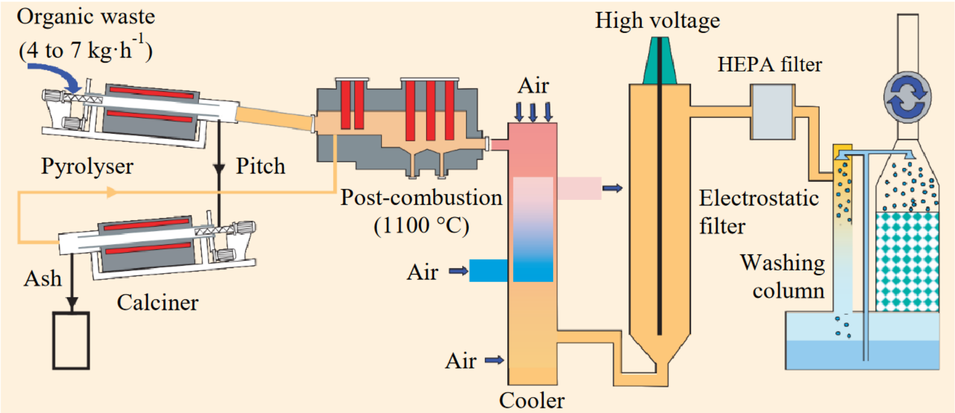

Figure 3: Diagram of the CEA’s IRIS process for the treatment of radioactive organic waste (Reproduced from [45]).

<u>Resource requirements:</u>

- Energy for heating (less than incineration).

Value assessment considerations

- H&S: Risk due to high temperature and the generation of combustible gases which need to be managed. Volume reduction may increase the concentration of contaminants and radionuclides in the ash.
- Risk and hazard reduction: Organics in the waste are destroyed.
- Environmental: Relatively lower energy cost as compared to incineration due to lower temperature.
- Socio-economic: Versatile, with a large range of potential feeds. Volume reduction results in lower storage and disposal costs.

Type of waste material that can be processed (input):

Suitable RSOW waste groups: [46,47]

- Spent IERs.
- Mixed organic waste, including halogenated plastics and cellulose.

Other wastes:

- Oily waste.
- Solvents.
- Sludge.
- Filters.
- Uranics.

Constraints and Exclusions:

Non-combustible waste is unsuitable. Halogenated plastics can be managed (as in the IRIS process) but require additional engineering.

Type of material produced by the process (output):

Pyrolysis itself produces tars and carbon rich char. In the context of radioactive waste management, an additional incineration or steam reforming step is typically applied after pyrolysis to increase the volume reduction and improve the wasteform[31,45,48]. The primary product in this case will be an inert ash. The composition of the ash is dependent on the starting material being treated.

Secondary waste:

Produces secondary gases which must be filtered and/or burned before discharge to the atmosphere. The filters and scrubbing fluid from the off-gas system must be replaced periodically and disposed of appropriately.

Is the product likely to need further conditioning prior to disposal?

Yes – the ashes will require further conditioning.

Current international practices:

Used in Europe, Japan, and the USA for the treatment of LLW and ILW (see below).

Facilities:

- Pyrolysis/incineration (Nyköping, Sweden)[49].
- Studsvik's inDRUM in-container pyrolysis technology (mobile)[50].
- IRIS process (CEA - Valduc, France)[51].
- Pyrolysis Resins in Mobile Electric installation (PRIME) is a mobile pyrolysis technology under development by Belgoprocess and Montair (mobile) [inactive pilot scale][48].
- Calcium Hydroxide Pebble pyrolysis reactor (Belgoprocess, Belgium) [Closed][31].

• 'Thermal Organic Reduction method (THOR)' (Tennessee and Savannah River, USA)[31]
Technical maturity: TRL 9 – Proven technology which is in commercial use for radioactive waste treatment.
Other comments: Catalysts may be used to increase the rate and efficiency of decomposition. Pre-treatment of the waste by size reduction, homogenisation or dewatering may be needed.
Work within PREDIS: CEA France has used the IRIS process to treat surrogate waste comprising of a mixture of organic solids and anionic IER to produce an ash. The first step to the IRIS process consists of oxidative pyrolysis at 550°C, producing pitch that is then processed in a calcining step at 900°C in an oxygen-enriched atmosphere. The pitch remains in the furnace for 2 hours and is transformed into ash mainly composed of Al, Si, Ca and Zn with a very low carbon content (< 1%).

3.2 Further Treatment technologies

The list of thermal treatment technologies available or currently under development suitable for the treatment of the RSOW has been extended beyond those studied in PREDIS WP6. The following datasheets are presented in this section:

- Acid digestion
- Supercritical water oxidation
- Thermochemical treatment
- Thermal desorption
- Vitrification

3.2.1 Acid digestion

Title: Acid digestion technology

Type of process: Treatment

Description of process:
Acid digestion is a technology that uses high temperature acid to oxidise the constituents of the feed material. It provides an alternative method of oxidising a waste in cases where incineration is not suitable, for example in the case of waste containing a significant amount of actinides or volatile radionuclides such as mercury and tritium[52].

Acid digestion can be adapted with different reagents and reaction conditions for different waste types. Of relevance, for radioactive mixed organic waste, is an acid digestion process developed by the US DOE for use at the Savannah River site. In this process waste is fed into a vessel containing nitric acid and phosphoric acid at a temperature between 130–200 °C (an earlier iteration of this process developed in Belgium used nitric acid and sulfuric acid at 250 °C). The nitric acid acts as an oxidant and the phosphoric acid acts as a carrier solution, preventing the nitric acid from boiling. Due to the carrier solution, the nitric acid remains in solution at atmospheric pressure and at temperatures needed for oxidation (> 130°C). When organic material is introduced into the solution, it is oxidised into CO_2, CO, and H_2O with a nitrogen oxide by-product. Inorganic constituents remain in solution and can be subsequently solidified in a glass or ceramic. Palladium is included as a catalyst at very low concentrations to enhance conversion of CO to CO_2 and N_2O to NO_x. It is possible to recover and recycle nitric acid from the N_2O and NO_x in the off-gas, which reduces the secondary wastes.

Resource requirements:

- Phosphoric and nitric acids.
- Palladium catalyst.
- Energy for heating (much less than incineration).

Value assessment considerations

- H&S: Risk due to the use of acids and high temperature.
- Risk and hazard reduction: Organics in the waste are destroyed.
- Environment: Relatively low off-gas volumes, production of nitrogen oxide which cannot be discharged and requires extensive off-gas treatment.
- Socio-economic: Volume reduction could greatly reduce interim storage and final disposal costs. Suitable for wastestreams with a significant content of volatile radionuclides.

Type of waste material that can be processed (input):

Suitable RSOW waste groups: [2, 52 ,53,54]

- Spent IERs.
- Mixed organic waste, including halogenated plastics and cellulose.

Other wastes:

- Filters.
- Munitions.
- Oil and Oily Wastes.
- Uranium or plutonium contaminated material.

Constraints and Exclusions:

Waste requires sizing (by e.g. shredding) prior to digestion for best efficiency. Treatment of halogenated organics is possible but results in the production of hydrochloric acid which requires additional handling. Bulk inorganic materials are not oxidised (digested) in this process but may be decontaminated.

Type of material produced by the process (output):

Acidic liquid residue containing inorganic constituents of waste (primarily as phosphates).

Secondary waste:

Spent scrubber solution and filters from off-gas system. If the NO_x is not recycled, then it must be managed as a secondary waste.

Is the product likely to need further conditioning prior to disposal?

Yes – liquid wastes require further treatment. Suitable methods include neutralisation of the acid solution followed by encapsulation in cement[2], or drying followed by immobilisation of residual solids in a glass or a magnesium phosphate ceramic wasteform[52].

Current international practices:

This review did not identify any currently operating acid digestion facilities for radioactive waste treatment. A pilot plant for the ‘Eurowetcomb’ acid digestion process to treat plutonium bearing wastes started operation in Belgium in 1980[55] and has since been decommissioned. The Savannah River site in USA subsequently developed the process to use a phosphoric acid carrier solution for organic in the 1990s. Acid digestion was used at the now shut-down Karlsruhe Reprocessing Plant (WAK) in Germany for the treatment of liquid reprocessing waste[56].

Facilities:

None identified.

Technical maturity:

TRL 9 (historically) – Industrial scale facilities have been operated for radioactive waste treatment, although none remain in operation.

3.2.2 Supercritical Water Oxidation

Title: Supercritical Water Oxidation / Hydrothermal Oxidation

Type of process: Treatment

Description of process:
Supercritical Water Oxidation (SCWO) or Hydrothermal Oxidation (HTO) is a thermal treatment process in which oxidation occurs in supercritical water. At pressures and temperatures above its critical point (P>22.1 MPa and T>374°C), water becomes a supercritical fluid; entering a state in which there are no distinct liquid and gaseous phases and the water has properties between those of its gas and liquid phases. Supercritical water acts as a very effective solvent and is completely miscible with oxygen and organic liquids, allowing for a very fast and complete oxidation reaction[57].

In a SCWO reactor, a mix of water, oxidant (typically an $N{:}O_2$ mixture or H_2O_2) and waste is subject to high pressure and temperature above the critical point of water which results in the spontaneous oxidation of the organics. This oxidation reaction occurs at a relatively low temperature (450-500°C) and results in an efficient conversion of organics to CO_2 with very little unwanted gaseous oxide production (SO_x and NO_x)[58,59]. Following discharge from the reactor, the supercritical mixture is cooled and allowed to expand, resulting in the separation of liquid and gaseous phases. The process products are a gaseous exhaust (CO_2, O_2, N) and an aqueous liquid effluent composed of water and salts.

The key technical challenges associated with this technology are corrosion and salt precipitation; a number of different SCWO reactor designs have been developed to manage these processes [58,60].

<u>Resource requirements:</u>

- Oxidiser ($N{:}O_2$ or H_2O_2).
- Water.
- Energy for heating (less than incineration).

Value assessment considerations

- H&S: Risks presented by high temperatures and very high pressures.
- Risk and Hazard reduction: Positive impacts due to destruction of organics and volume reduction.
- Environment: Lower energy requirements than incineration. Has a relatively ‘clean’ gaseous exhaust.
- Socio-economic: Bespoke facility required, although with a potentially small footprint, which can fit inside a glovebox[57]. Lower storage and disposal costs due to reduction of waste volume.

Type of waste material that can be processed (input):
Suitable RSOW waste groups:

- Spent IERs.
- Mixed organic waste.

Other wastes:

- Organic liquids.
- Solvents.
- Filters.

Constraints and Exclusions:
Unsuitable for inorganic waste. Solid waste must be size reduced (i.e. shredded).

Type of material produced by the process (output):

- Liquid effluent (H_2O, salts).

Secondary waste:

- Gaseous exhaust (CO_2, O_2, N).

Is the product likely to need further conditioning prior to disposal?
Yes, the liquid effluent requires further treatment (some form of dewatering and immobilisation) to be suitable for disposal.

Current international practices:
A number of SCWO plants have been built for commercial treatment of non-radioactive wastes[61].

The Los Alamos National Laboratory in the USA have used this technique for the treatment of alpha-contaminated waste including solvents, rags, filters and IERs [2].

The CEA operate the 'DELOS' facility, a compact HTO process for radioactive liquid organic effluents[57,60]. This is implemented as a small continuous feed organic effluent treatment facility at the Marcoule site. This facility can theoretically treat organic solids, although this is still experimental.

Facilities:

- DELOS facility at Marcoule (France).

Technical maturity:
TRL 6 – SCWO has been implemented for the treatment of radioactive liquid organic waste, although there has only been pilot scale treatment of solid radioactive organic waste.

3.2.3 Thermochemical treatment

Title: Thermochemical treatment/Self-propagating high-temperature synthesis

Type of process: Treatment

Description of process:

Thermochemical treatment involves mixing waste with a Powdered Metal Fuel (PMF) which chemically reacts with itself and the waste to achieve very high temperatures[31,1]. PMFs generally consist of a powdered metal (such as aluminium or magnesium), an oxygen-containing component (such as iron oxide), and some additives. The PMF is usually formulated to react chemically with the waste or components of it and to capture and immobilise radionuclides and any hazardous chemical species.

The product of thermochemical treatment is dependent on the specifics of the waste and PMF. For treatment of organics on their own, the product is typically a mixture of ash and slag. Treating inorganic materials, or adding glass forming materials such as sand, results in a glass/ceramic product[62,63].

Resource requirements:

- Metal powder, combustion activator and additives.

Value assessment considerations

- H&S: Risks due to high temperatures, the handling of reactive substances (the PMFs), and the production of flammable gases (hydrogen).
- Risk and hazard reduction: Destruction of organics. Process may be designed to produce a stable wasteform.
- Environment: Significant embodied energy costs in the production of metal powder. Corrosive gases are destroyed in the reaction.
- Socio-economic: Volume reduction resulting in low transport and storage costs. Treatment can be done with little or no fixed plant.

Type of waste material that can be processed (input)[2]:

Suitable RSOW waste groups:

- Spent IERs
- Mixed organics

Other wastes:

- Metals.
- Concrete.
- Contaminated soil.
- Filters.
- Uranics.

Constraints and Exclusions:

Unsuitable for volatile radionuclides that may evolve into gas at high temperatures. The treated waste (i.e. slag and ash) may present challenges for conditioning to a disposable form.

Type of material produced by the process (output): For organics, an ash-slag mixture is expected.
Secondary waste: Off-gas treatment required for organic wastes (with associated secondary waste).
Is the product likely to need further conditioning prior to disposal? Yes – Ash and slag likely to require immobilisation.
Current international practices: Thermochemical treatment has been used on a pilot scale in Russia and the Czech Republic for surface decontamination and for the treatment of IERs, animal cadavers, contaminated clay, reactor graphite and irradiated zirconium[31]. The process is also being studied in China for the immobilisation of contaminated soil, plutonium containing waste, and graphite[62]. Facilities: None identified.
Technical maturity: TRL 6 – Has been implemented for the treatment of radioactive organic waste at a pilot scale.

3.2.4 Thermal desorption

Title: Thermal Desorption

Type of process: Treatment

Description of process:
Thermal desorption consists of heating a contaminated material to evaporate contaminants, which are then recovered or destroyed[64,65]. To be a viable treatment, contaminants must evaporate at a lower temperature than the bulk waste. Contaminants that may be removed with thermal desorption include volatile metals such as mercury and Volatile and Semi-Volatile Organic Compounds (VOCs and SVOCs) (which include polychlorinated biphenyls (PCBs), pesticides, and various petroleum products). The temperature of the thermal desorption process is generally selected based on the boiling point of the contaminant(s) being removed; for VOCs, thermal desorption occurs at temperatures of between 200 and 400°C, for SVOCs, higher temperatures of up to 550°C may be needed[64,66]. The boiling point of volatiles may be lowered by reducing the pressure in the treatment vessel; in the variant process Vacuum Thermal Desorption (VTD), the waste is held under vacuum for treatment.

For radioactive waste management, two potential applications of thermal desorption could be considered:

- The removal of conventional hazardous chemicals (VOCs, PCBs, mercury, etc) from radioactively contaminated wastes[64]. The radioactive product will in this case be the same as the initial wasteform but with volatiles removed, typically allowing for further treatment and disposal.
- The decontamination of a waste containing volatile radionuclides (for example detritiation of some materials)[67,68]. The radioactive product in this case would be the separated contaminants.

<u>Resource requirements:</u>
- Energy for heating (much less than incineration).

Value assessment considerations
- <u>H&S:</u> Risk due to volatilisation of hazardous chemicals and/or radionuclides.
- <u>Risk and hazard reduction:</u> Reduced handling disposal and storage risks due to removal/separation of hazardous chemicals or chemicals impacting disposability (if using for removal of hazardous chemicals).
- <u>Environment:</u> Fairly low energy use compared to other technologies. Bulk decontaminated material may be suitable for reuse or release (if using for decontamination).
- <u>Socio-economic:</u> The decontamination of the wasteform can potentially open additional management routes.

Type of waste material that can be processed (input):
To be a viable treatment, contaminants must evaporate at a lower temperature than the bulk waste; contaminants that may be removed with thermal desorption include volatile and semi-volatile organic compounds (which include PCBs, pesticides, and various petroleum products) and volatile metals such as mercury.

Type of material produced by the process (output):

The primary (largest volume) product is the decontaminated waste, which can be expected to be chemically the same as the pre-treatment waste, but dried, and with the volatiles removed. Depending on the process application, this waste may be radioactive or clean.

Secondary waste:
Recovered evaporated volatile contaminants. Final form is dependent on the contaminant and treatment approach. Most evaporated contaminants can be cooled and condensed, resulting in separated liquid or solid volatiles which will likely need subsequent treatment. Organic volatiles may be destroyed by combustion while still in a gas phase, leaving a small volume of residue.
Secondary wastes will also arise from filters / scrubbers from the off-gas system.

Is the product likely to need further conditioning prior to disposal?
Waste dependant: the main product by volume is the decontaminated bulk material which may either be suitable for clearance or may require subsequent conditioning for disposal. If there is activity in the secondary waste, this will need conditioning prior to disposal.

Current international practices:
Thermal desorption is commonly used in non-radioactive contexts to treat soil contaminated with VOCs and SVOCs [65]. It has also been adopted for the removal of conventional contaminants from radiologically contaminated soil, sludge, and liquids arising from US DOE site clean-up and remediation activities[64,69].

The Materials Detritiation Facility (MDF) at Culham, UK uses thermal desorption for removing tritium from non-combustible tritium contaminated waste from the JET Fusion experiment[68].

Bench-scale experiments have been undertaken in Korea to demonstrate the removal of tritium and C-14 from activated carbon[67] by thermal desorption.

<u>Facilities:</u>
- MDF at Culham (UK).

Technical maturity:
TRL 9 – Proven technology which is in use for radioactive waste treatment.

3.2.5 Vitrification

Title: Vitrification

Type of process: Treatment/conditioning

Description of process:
The vitrification process involves melting a material in a high temperature furnace, then cooling it to form a glass[70]. Vitrification has a long history of use for immobilising highly active liquors from reprocessing.

There are a number of technologies for creating the waste melt which are suitable for an array of waste types and activities. At a high level, vitrification technologies can be classified by their heating method, which typically consists of either induction or joule (resistance) heating. Heat may be transmitted directly to the waste itself (direct heating) or via a heating element (indirect heating). Vitrification technologies can be further distinguished by the details of the crucible in which the waste is melted; key characteristics include the crucible material (metal or ceramic), whether the crucible is hot- or cold-walled and whether the crucible forms part of the disposal package (i.e. in-package melting)[71]. In-situ vitrification of, for example, contaminated soils is also possible.

A calcination or incineration step is typically required for organic and liquid wastes to create a dry powdered product suitable for vitrification[72]. This step may be integrated into the vitrification process as a distinct step (as in, for example, the Atelier de Vitrification La Hague (AVH) process) or may occur during the heating of the waste into a melt (e.g., GeoMelt®[73], SHIVA). This step will require some form of off-gas handling.

The vitrification feed is mixed with a glass former, usually a borosilicate, as these have a relatively low melting temperature and the resulting glass has good properties. Once mixed, the product is heated to melt the waste and glass former. The melted waste can then be poured into a disposal container or allowed to solidify in-situ. The product is a mechanically and chemically durable glass suitable for storage and disposal [74].

Resource requirements:

- Energy for heating (much more than incineration).
- Glass forming additives.

Value assessment considerations

- H&S: Risk due to very high temperatures.
- Risk and Hazard reduction: Organics are destroyed, the product is a stable, solid wasteform suitable for storage or disposal.
- Environment: Very large amount of energy is required in comparison to incineration. Silicate wastes such as glass fibre filters or asbestos may be co-processed as glass-formers.
- Socio-economic: Potentially quite versatile. Volume reduction can reduce disposal and storage costs. High capital and operating costs.

Type of waste material that can be processed (input):
Suitable RSOW waste groups:

- Spent IERs [70].
- Mixed organic waste.
- Conditioned waste, including cemented waste.

Other wastes:

- Filters.
- Sludge [75].
- Inorganic waste.
- Highly active liquour (following calcination).
- Metals.

Constraints and Exclusions:

Unsuitable for waste containing volatile radionuclides or waste which will not melt at high temperatures (e.g. graphite). Likely to be un-economical for large volumes of low activity waste. For RSOW, an additional incineration/calcination step is typically required prior to vitrification.

Type of material produced by the process (output):

Homogenous glass characterised by good mechanical strength and chemical durability.

Secondary waste:

Off-gas handling required for organic or aqueous waste.

Is the product likely to need further conditioning prior to disposal?

No – vitrification produces a durable and stable wasteform suitable for interim storage and final disposal.

Current international practices:

Vitrification has been widely used in a non-nuclear setting for treating sewage sludge, as an alternative (or in conjunction with) incineration [76,77]. The vitrified product results in a glassy waste which can be re-used as aggregate, or (if of high enough grade) as insulation material.

In the nuclear industry, vitrification is used to treat waste in the nuclear industries of France, Russia, Japan, Slovakia the UK and the USA [70,78]. There are a wide range of vitrification technologies using various melters. The AVH process is in use in France and the UK for vitrifying high level waste from reprocessing. The GeoMelt® In-Container Vitrification technology has been deployed for radioactive waste treatment in USA, UK, Japan and Australia. The DEM'N'MELT in-can vitrification process is under development (at the pilot stage) in France[74].

Facilities:

Industrial scale (active facilities by melter type)[79]:

- Cold Crucible Induction Melter (CCIM) (La Hague, France and Ulchin, S. Korea).
- Joule-heated ceramic melter (JHCM) (Savannah River and Hanford, USA).
- Induction, Hot credible (IHC) (Sellafield, UK)
- GeoMelt® (mobile).

Trial scale:

- Advanced-CCIM (CEA Marcoule, France).
- DEM'N'MELT (CEA Marcoule, France).

In addition, there are a number of plants using plasma technology in conjunction with vitrification (see plasma arc processing sheet).

Technical maturity:

TRL 9 – Proven technology which is in use for commercial radioactive waste treatment.

4 Conclusions

The reviewed thermal technologies are summarised in Table 4.1, which identifies the TRL and process product of the technology and provides a short summary of the key features of each technology.

Table 4.1: Summary of the reviewed thermal technologies.

Technology	TRL	Process product	Features
Gasification	6	Ashes	Significantly less unwanted SO_x and NO_x than is produced by incineration. Product requires conditioning. Unsuitable for wastes containing significant quantities of volatile radionuclides.
HIP	7	Glass-ceramic or ceramic	Require thermal pre-treatment step. Good containment throughout process producing a stable ceramic wasteform.
MSO	6	Spent salts	Acidic gases retained in salt. Relatively low operating temperatures and clean off-gas relative to incineration. Product requires conditioning.
Plasma	9	Slag or glass	Can accept a wide range of unsorted raw waste. High temperature and operating costs. Produces a stable, disposable product.
Wet Oxidation	7	Inorganic sludge	Contaminants retained in solution resulting in a clean off-gas, and low overall capital cost. Product requires conditioning.
Pyrolysis	9	Ashes	Lower operating temperatures than incineration, allowing the retention of relatively more volatile radionuclides. Product requires conditioning.
Acid Digestion	9	Acidic sludge	Low temperature and relatively good retention of radionuclides. Significant off-gas management required. Solid waste requires pre-treatment (shredding). Product requires conditioning.
SCWO	6	Inorganic sludge	Very clean gaseous exhaust. High capital cost. Solid waste requires pre-treatment (shredding). Product requires management.
Thermo-chemical	6	Slag, ash	High temperatures and very poor retention of volatile radionuclides. Low capital cost.
Thermal Desorption	9	Waste decontaminated of volatiles	Only suitable for decontaminating waste containing volatile radionuclides or volatile organic or inorganic contaminants.
Vitrification	9	Glass	Versatile treatment producing a very stable wasteform. Volatile radionuclides not retained.

The reviewed thermal treatment technologies have varying compatibility with different types of RSOW. The compatibility of different types of RSOW with the reviewed thermal treatment technologies is rated according to a Red, Amber Green (RAG) system, where:

- **Green** Technology is applicable for this waste type.
- **Amber** Technology is potentially applicable for this waste type, although may not have been demonstrated.
- **Red** Technology is incompatible with this waste type.

The RAG ratings for each technology/RSOW combination are presented in Table 4.2.

Table 4.2: Matrix of the RAG ratings for the compatibility of treatment and conditioning technologies with RSOW wasteforms.

Technology	IERs	Mixed Organic	Conditioned waste		
			Bituminised	Polymeric	Cemented
Gasification	Green	Green	Amber	Amber	Red
HIP	Green	Green	Amber	Amber	Red
MSO	Green	Green	Amber	Amber	Red
Plasma	Green	Green	Green	Green	Green
Wet Oxidation	Green	Green	Amber	Amber	Red
Pyrolysis	Green	Green	Amber	Amber	Red
Acid Digestion	Green	Green	Amber	Amber	Red
SCWO	Green	Amber	Red	Red	Red
Thermochemical	Green	Green	Amber	Amber	Amber
Thermal Desorption	Amber	Amber	Red	Red	Red
Vitrification	Green	Green	Green	Green	Green

This review has summarised the types of RSOW requiring treatment as well as the thermal treatment and conditioning technologies suitable for the treatment of RSOW that are available or under development. This analysis may be used by waste owners and national waste management programmes to inform an options assessment for the management of RSOW. PREDIS Deliverable 6.3 provides an example of such an assessment, undertaken at a generic level and focusing on a subset of the RSOW treatment technologies studied within PREDIS.

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