Thermal Processes for Immobilising Intermediate Level Wastes

Position Paper

Thermal Processing

Thermal Processes for the Immobilisation of Intermediate Level Wastes

The UK has gained experience of a number of processes for the immobilisation of radioactive waste over several decades. This experience comprises development of product formulations, immobilisation processes, as well as commissioning, operating and optimising immobilisation plants and includes innovations that have been adopted internationally.

Since 1991, high level liquid waste from reprocessing has been vitrified using a French process for inductive heating of calcined liquid waste, whilst most operational intermediate level wastes have been directly encapsulated in cement. Although substantial progress has also been made in the vitrification of high level wastes produced before 1991, there remain a number of legacy intermediate level wastes from these earlier days of reprocessing for which decisions on immobilisation have yet to be made.

Thermal processes have the potential to provide high quality glass and ceramic based waste products and may offer significant savings on operational and disposal costs of these wastes due to the potential for reduced volume in comparison with cementation. A wide range of thermal processes exist worldwide that could be used for immobilisation of some of these UK legacy wastes.

This paper discusses the potential benefits of thermal processes as well as the challenges that would need to be overcome to implement thermal processes for intermediate level wastes. The UK's track record in operating Waste Vitrification Plant is discussed along with a range of other technologies which could be applied for ILW.

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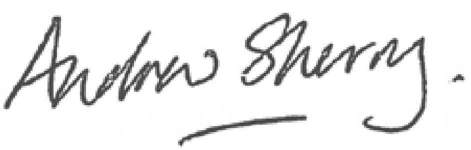

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Introduction

In the UK, significant progress has been made in the immobilisation of High Level Waste (HLW) and Intermediate Level Wastes (ILW) from the nuclear fuel cycle while Low Level Waste (LLW) is processed for disposal in an operational Low Level Waste Repository (LLWR).

The application of thermal treatment processes for immobilisation of UK wastes has so far been restricted to HLW vitrification, whereas ILW has generally been immobilised within a cement matrix. With over 20 years UK operational experience for a wide range of waste streams, cementation has also been selected as the baseline technology for most of the legacy wastes that still require immobilisation.

The benefits of cementation are that the process is simple, employing a readily available and low cost material that is compatible with wet wastes and operates at low temperatures. Waste pre-treatment stages and secondary waste generation can thus be minimised or avoided. Furthermore, extensive operational history and academic research has generated a considerable body of knowledge and experience which tends to favour its selection for future waste treatment plants.

Yet the selection of cementation has required compromise. The absence of pre-treatment stages has meant that waste streams such as reactive metals or organic plutonium contaminated material (PCM) retain reactivity within the product resulting in additional considerations during storage and disposal. Wastes with a low bulk density or high water content tend to have low waste loadings, and even where high waste loadings are possible, plant operational factors often result in the process operating at sub-optimal waste loadings.

Thermal treatment processes offer the potential to overcome these disadvantages for a range of wastes. Thermal technologies are those which through the application of heat can effect the immobilisation of radioactive elements in an inorganic, chemically inert and largely homogenous wasteform, breakdown organics and, in some cases, oxidise metallic components. This includes vitrification, ceramic immobilisation, metals melting and incineration, as well as a host of others which could form the basis of a future strategy for immobilising ILW in the UK.


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Thermal Treatment Processes

Benefits of Thermal Treatment Processes

With such a wide range of thermal technologies, the potential benefits compared to baseline technology could include:

Removal/reduction of reactivity

This can be achieved for example by oxidation of small amounts of reactive metal or by melting and phase separation to reduce metal surface area. Whilst the Geological Disposal Facility (GDF) is designed to manage waste products containing reactive metals, this could reduce the need for monitoring during interim storage and remove a possible requirement for rework before transport and disposal.

Destruction of organic material

There are several advantages from organics destruction. Soluble degradation products of materials such as cellulose have the potential to increase solubility of actinides. Non-aqueous phase liquids may also be formed which could provide a transport pathway for a range of species.

Volume Reduction

The waste volume can be reduced relative to unconditioned waste by removal of water or oxidation of bulky organic wastes such as PCM. Furthermore, the mineralogy of the product can potentially be formulated so that the waste itself is an intimate part of the immobilising matrix. For example, the open structure of inorganic ion exchangers can be collapsed into a higher density mineral with little or no addition of other materials.

Increased waste loading is potentially the most significant benefit of thermal processes, since it would reduce the operational lifetime of the encapsulation plant as well as the number of waste products requiring storage over many decades, then transport to a GDF. Carried out on a sufficiently large scale, it could also reduce the volume of GDF excavation. Together, these benefits offer major opportunities for reducing processing and disposal costs.

Product Quality

The Waste Package Specifications published by RWM Ltd [1] provide a set of requirements to be met by waste packages which will be subject to geological disposal and which therefore must be considered when designing a waste immobilisation process and product. The products of thermal processes offer improved performance relative to cements for several of these criteria, including immobilisation of radionuclides (through lower porosity, permeability and leachability) [2], chemical containment (through destruction of various materials of concern, e.g. complexants) and wasteform evolution (through minimisation or removal of reactivity).

Challenges For Thermal Treatment Processes

Set against these potential benefits there are a number of challenges that will need to be overcome before thermal processes are adopted for UK ILW.

Radioelement volatility

Tritium, carbon-14 and iodine are difficult to contain unless a closed system such as Hot Isostatic Pressing (HIP) is used. The extent of the challenge for other radioelements will depend on various factors such as operating temperature or composition of the glass forming additives. For instance, the fraction of Cs which is reported to be volatilised during vitrification varies widely.

Process complexity

Operation at high temperature introduces additional process complexity, so that innovative approaches will be required to minimise in-cell components requiring maintenance.

Waste and waste product heterogeneity

To some extent, the current waste treatment plants have dealt with the easier waste streams, since operational waste promptly processed is well characterised and reasonably homogeneous. Legacy wastes on the other hand are often a poorly characterised mixture of components with the potential for uncertainties.

Development of formulation envelopes

To ensure product quality, it is necessary to establish a "formulation envelope", which is the compositional range within which an acceptable waste form will be produced on plant. This introduces a need for significant research and development on each waste stream and during plant operation, quality control procedures to monitor waste composition and the formulation of additives. To guarantee product quality during plant operation and to take account of variability in waste composition for instance, the formulation envelope will not usually extend to the maximum waste loading that has been shown to be achievable during research and development. The claims for high waste loadings for thermal treatment processes therefore need to be supported by extensive R&D evidence taking account of these issues.

Development of process envelopes

Waste immobilisation plants are operated in accordance with a quality assurance system which is based upon:

- The specification of all the parameters which determine the characteristics of the encapsulated product or reflect on the plant operability.
- The specification of the envelope of these parameters which process development work has shown will guarantee a satisfactory product within plant operating constraints.
- The ability of the process equipment and controls to ensure that the parameters are maintained within the specified envelope.

A thorough understanding of these parameters is required in order to establish a "process envelope" to ensure production of a quality product. The challenges for ILW immobilisation in developing a process envelope can be illustrated with reference to current operational plants. Both the Waste Vitrification Plant (WVP) described below and the current range of cementation plants [3] have established operational procedures to manage key process parameters that have the potential to affect the quality of the product. Over and above the underpinning work associated with the development of waste product formulations, there is therefore a need to develop process envelopes for thermal processes. This topic also introduces an opportunity for further innovation, with the development of in-line process monitoring and control, for instance for the measurement of temperature, mix composition and off-gas analysis.

Thermal processes have so far not been adopted in the UK for ILW and international experience has been limited, particularly in terms of the activity of wastes processed. Nevertheless, substantial experience has been gained through the development and operation of the WVP for high level liquid waste (HLW). The approach taken and the benefits gained provide an excellent model for the development of thermal processes for ILW immobilisation.

HLW Vitrification Experience in the UK

The aqueous raffinate from fuel dissolution and subsequent solvent extraction is evaporated and stored as Highly Active Liquor (HAL) prior to vitrification. HAL from all previous and existing reprocessing plants is a solution of metal nitrates in nitric acid that also contains some insoluble solids. The feed to the Waste Vitrification Plant (WVP) is a well-mixed blend of one or more of these liquors fed from a tank that has been sampled and analysed. The composition of the feed tanks vary within a known range and during operation of WVP the waste feed is homogeneous and of a well characterised composition.

Development work to substantiate product quality for WVP was performed at a range of scales from 50g laboratory melts to full scale and included substantiation of the development work during commissioning of the active plant using non-active HAL simulants [4,5]. Significant small scale laboratory studies were undertaken to assess general compositional variation, eg blend variation, and specific key element studies, including enhanced levels of iron, chromium, nickel, molybdenum, sulphur and sodium from fission products, corrosion products and process additives. Experiments were performed at a range of waste loading levels, temperatures and degrees of mixing to identify the limits of the process and operational envelopes. Products were characterised to confirm they were fully reacted and homogeneous, without the formation of separate phases. These were tested for chemical and thermal resistance and other physical properties.

Pilot scale and large scale testing was then performed under conditions that produced acceptable products in the laboratory. Operational parameters such as feed rate, temperature, mixing in the melter were varied to assess the impact on the glass product and the operability of the process equipment. Several operational campaigns were undertaken prior to the commissioning of WVP to investigate the intermediate calcine products using a range of

Figure 1: *NNL's Vitrification Test Rig supports the UK HLW Waste Vitrification Plant at Sellafield*

feed types and blends. Optimisation of the plant parameters required a number of experimental campaigns. Long term stability testing for process performance of feed and off gas systems was performed and key element studies were reassessed at large scale and some variation in the waste loading was tested [6].

Following several years of operation of WVP a full scale test rig was constructed in 2004 to help to improve the plant's performance and flexibility. The Vitrification Test Rig (VTR) replicates the feed systems, main process equipment and primary off gas system of WVP. The original development work for WVP had relied on theoretical waste composition data. The actual fission product content of the waste from Oxide reprocessing was lower than the theoretical flowsheet and provided an opportunity to increase the waste loading in the product glass. The VTR was necessary to confirm product quality under all representative operational conditions on WVP. The main aims of the VTR were to improve waste loading, increase liquor feed rate, glass production rate and plant availability, to confirm product quality of glass made from future feeds and qualify products made on WVP outside the previously specified operational envelope. It has completed all of these tasks satisfactorily [7-14].

Examining the waste loading a little more closely, the VTR was first set up to operate in the same way as WVP, at the same feed rate and waste loading (using simulated HAL). The performance of the plant was assessed and compared with WVP data to confirm the similarity in their behaviour. The waste loading was increased in increments and tested at a range of glass production rates over the full operational envelope of WVP. This was then extended further to provide more latitude for the operators at WVP, resulting in more flexible operations and less likelihood of producing an out of specification product. All waste types and blend ratios were assessed and using a melter with an improved mixing system a new waste loading limit was produced with significant headroom to accommodate sampling, analytical and process feed variation. The waste loading limit has been raised from 25%w/w to 35%w/w and has been implemented on WVP at up to 34%w/w to date.

The decision to construct the VTR followed a review of HAL strategy in which it became clear that a 4th vitrification line costing £400 million would be required to meet the HAL stocks reduction programme unless a step change in throughput could be achieved. The VTR has provided operational data to support increased HAL throughput and together with equipment improvements obtained from Areva, this expense was avoided.

Additionally, savings on vitrified product numbers of over 200 containers have been achieved and future savings will be significantly higher. The production and disposal cost for each container is over £500k and the savings made on WVP to date have more than paid for the capital and operating costs of the VTR with considerable future cost benefit being delivered.

The systematic approach taken during the development programme for WVP can be applied to alternative technologies used for vitrification of ILW, but the heterogeneity and variability of the ILW feed will require greater attention. The importance of scale was evident on the work performed for HLW. The achievable waste loading on the VTR was greater than for small laboratory melts, possibly as a result of longer melt duration and better mixing. Limiting the production rate or waste loading of ILW products by inadequate scale testing must be avoided if maximum benefit is to be realised.

Other Wastes Suitable for Thermal Processing

Legacy wastes have arisen from historic processing and long storage periods and pressing operational needs in the 70s and 80s have led to poorly characterised material often significantly corroded during interim storage. These wastes contain a variety of components making them difficult to treat. Irradiated fuel and uranium residues are present along with activated and contaminated metals. Depending on the length of time they have been stored, these may have undergone varying extents of corrosion and dissolution leaving previously solid material in the form of sludges of uncertain composition. Other legacy materials consist of flocs, graphite and ion exchange resins; all require specific solutions.

Processing of plutonium leaves a legacy of contaminated consumables and equipment which due to the radiotoxic nature of plutonium require a particular approach. Pu contaminated material (PCM) contains any materials having been in contact with plutonium and as such are varied and generally contain a variety of organic material such as Personal Protective Equipment, and structural material such as steel and concrete. This variety makes treatment complex. Currently PCM is treated through a compaction and cementation process but the existing plants may have insufficient capacity to deal with the large quantities of PCM which will arise when facilities are decommissioned and during operation of plant required to manage ultimate Pu disposition.

Future decommissioning and decontamination of nuclear facilities will give rise to high volumes of metals and concrete which will require treatment. Methods for reducing the volume of material assigned to repositories, and for increasing possible clearance as non-radioactive will be required eg for re-use in the industry.

Future UK electricity generation scenarios include options for much higher nuclear generating capacity than at present, including options for a closed fuel cycle based on either aqueous or pyrochemical recycling. Waste volumes will increase and storage and disposal capacity will need to be managed. Waste loadings must ideally be higher than currently achieved despite higher radioactivity concentrations. Thermal treatment processes are a potential solution for both aqueous and pyrochemical processing.

"Thermal treatment processes are a potential solution for the management of plutonium contaminated material."

Technology Options

There are various examples of thermal treatment systems either in current use or under development internationally for radioactive waste immobilisation [15]. Particular examples are described below.

Joule heating

The vitrification of radioactive wastes requires raising the temperature of blended feeds in order to break chemical bonds and facilitate reactions between species. Controlled cooling through the glass transition temperature then facilitates the formation of a glass in which the majority of radioelements are intimately bonded as part of the glass structure with the remainder being encapsulated within the glass matrix as separate phases.

The majority of melters used for this process rely on joule heating where energy is directly imparted to the melt through the use of electrodes. At room temperature frit/waste feed blends are non-conducting and a starter path is required to raise the temperature to the level at which the mix becomes conducting and energy can be transferred into the mix producing a glass melt. Typically these operate at temperatures in excess of 1100˚C. Waste loadings may vary from around 25 wt%, more typical of HLW vitrification, to almost 100 wt% where careful blending of feeds can provide all the glass forming species required in order to form a melt.

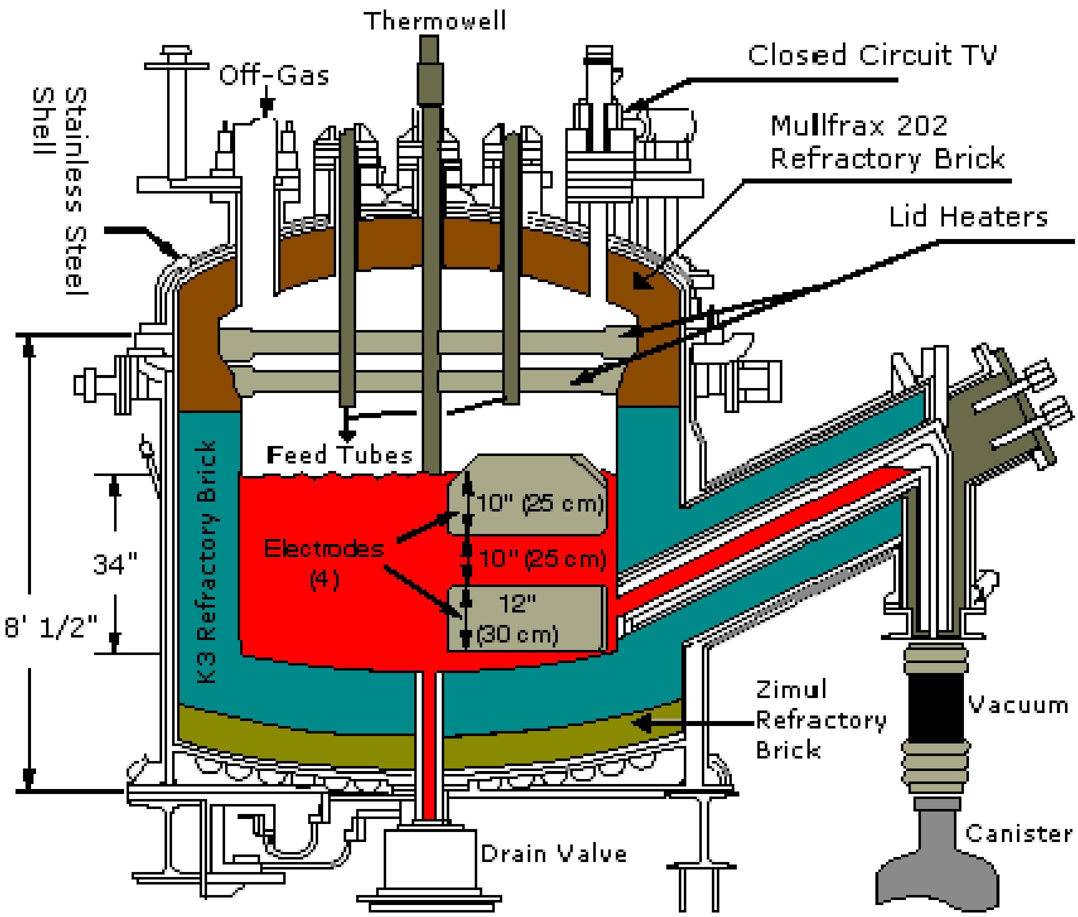

Figure 2: *Schematic cross section of a Joule heated ceramic melter*

Technology Options continued

In these systems once the feeds have been melted and conditioned in the melter they are poured into canisters and cooled ready for consignment to interim storage and disposal. Typically these systems will be operated on a continuous basis with frit and wastes being fed onto a cold cap on the top of the melt. This feed is then assimilated into the melt.

The joule heated system is the most commonly deployed melter system and has been successfully operated on USDoE sites, such as West Valley and Savannah River for the vitrification of reprocessing wastes in Germany and Belgium where melters have completed their missions. They are also deployed in Russia, eg Mayak and at reprocessing facilities in Japan.

A typical example of a Joule Heated Ceramic Melter is shown in figure 2. The glass pool is heated by submerged electrodes. The waste and frit are fed through the roof of the melter and bubblers can be used to augment mixing in the melter thus promoting reaction between feed and frit, contributing to a homogenous final product.

The main technical challenge specific to such continuous melting joule heating systems is the pouring system. For example with HLW, high levels of noble metals can block bottom pouring systems: induction heated bottom pouring vavles can be easily blocked through the settling of noble metals in the glass melt. Airlift systems can mitigate this issue but have complexities of their own. Failure to remove noble metals from the system can result in build up and potential shorting of the electrodes and reduced energy transfer to the melt. Bubbling systems can enhance mixing and prevent noble metals build up.

Wasteform development is important in ensuring digestion of waste into a melt with the required viscosity characteristics to promote adequate mixing and allow pouring. Addition of boron for example can increase the ability of the glass to digest waste components but has the potential downside of creating more volatile components, thus putting a greater load on the off-gas system. Non boron systems can potentially operate at higher temperatures with lower radioelement volatility.

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Joule heating ICV (In-Container Vitrification)

A variant of the joule heated system is where the melter doubles as the containment system. Early developments at UKAEA Harwell and CEA Marcoule investigated such systems for the vitrification of high active wastes [16] but throughput was considered an issue and such developments were dropped in favour of the calciner/melter 2 stage process (AVH system).

Such an ICV system, known as Geomelt, has been latterly developed from initial work on in situ vitrification where clean-up of Pu-containing soils was demonstrated at the former nuclear bomb test site in Maralinga in South Australia [17]. The potential for ICV to treat a range of UK ILWs has also been evaluated [18]. The avoidance of the

Figure 3: *ICV Melter, Hanford (courtesy of Kurion)*

need to pour in this system gives flexibility in the choice of glass compositions. The process is by definition a batch one and variants of how the melter is operated can be used to suit differing waste types. Pre-emplacement of wastes in the melter cavity can be carried out to enable treatment of miscellaneous solid wastes. The resulting wasteform can be homogeneous, akin to HLW glass, but for lower activity wastes, complete homogeneity may not be necessary to meet disposal requirements, thus potentially reducing costs of processing.

The ICV system and continuous joule heated systems differ only in the lack of a pouring requirement with the former. Challenges to the off gas system for example are largely equivalent. The ICV technology is operated in a batch mode by its very design and so by comparison to the continuously operated joule heated systems throughput could be a challenge. Although bubblers designed to increase throughput could be incorporated, the utility of the system lies in its simplicity which may be compromised by additional components. The key challenge therefore is to combine intelligent melter operation with appropriate glass composition to deliver optimum processing.

Technology Options continued

Cold crucible

'Cold crucible' is a term given to melter systems where the containment is water cooled. The cooling prevents the outer layer of feed/frit from melting and as such the melt is carried out within a 'skull' of unmelted feed/frit. This prevents adherence of materials to the wall and thus corrosion and decontamination is reduced. Such a skull also allows the operation of higher temperature melts which would normally be in excess of the melting point of the containment material.

A key practitioner in this field is the French group Areva who operate cold crucible melting at their La Hague HLW vitrification facility [19]. Its implementation has allowed the facility to process wastes requiring more refractory glass compositions and has been shown to enhance throughput for the same melt surface area.

A similar technology is operated by Radon SIA in Russia for the treatment of lower activity wastes [20]. The cold crucible systems have throughputs of around 25-50 kg/hr and so may be best suited to homogeneous waste streams of high activity or low volume niche wastes.

To date, cold crucible melter systems have largely been employed on homogeneous liquid fed wastes. They have also been limited in size largely due to the requirement of maintaining a consistent induction field in the glass melt. While throughput over the corresponding hot wall induction melter is much improved it remains to be seen whether such technology would be appropriate for miscellaneous feeds containing appreciable amounts of solid components. However, due to the cold wall technology these melters could prove ideal in dealing with corrosive feeds that require higher temperature processing.

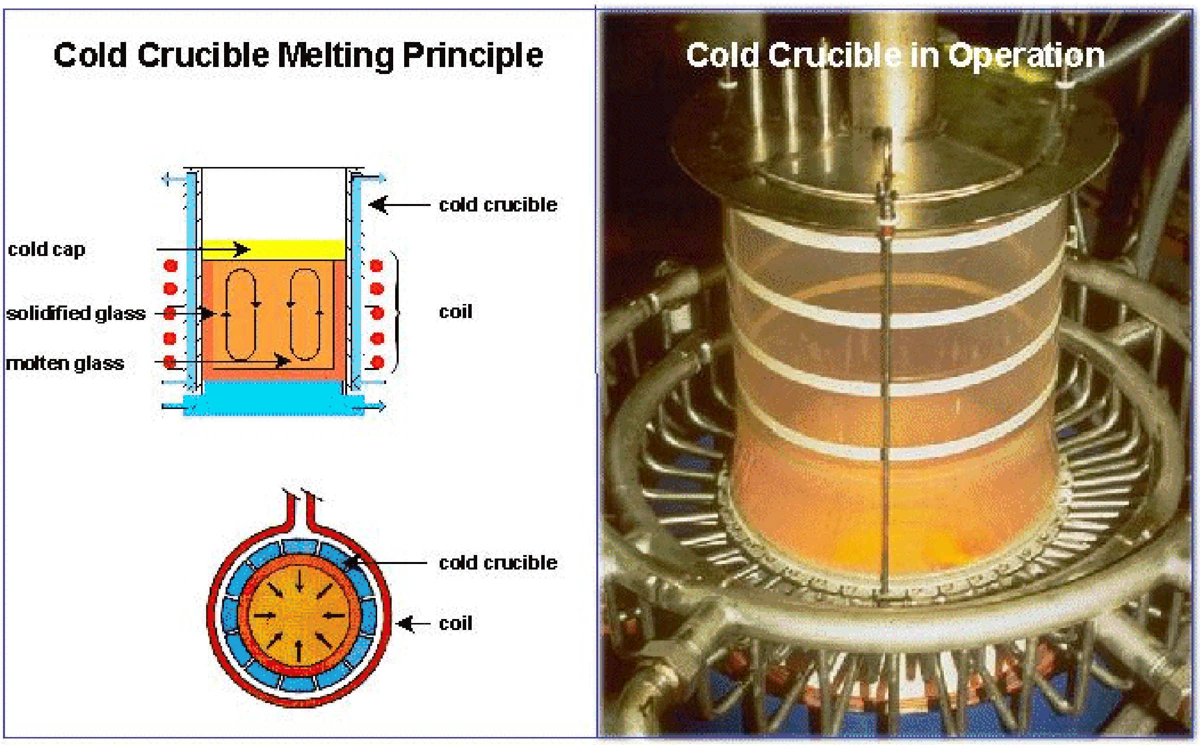

Figure 4: *Principle of operation and top view of cold crucible melter (courtesy of CEA)*

Plasma treatment

Energy is imparted to the feeds and a melt is produced either by a transferred arc (electrode to melt pool) or a non-transferred arc (electrode to electrode).

The use of plasma to transfer energy into melts has been quite extensive in the treatment of municipal waste, not only to passivate hazardous components but to produce gases as a by-product which have in turn been used to produce electrical energy.

The very high temperatures deployed in plasmas provide for the complete dissociation of organics but do require extensive off gas treatment facilities. An example of plasma processing and the evolution of a conventional plasma plant for radioactive wastes is seen at the Zwilag facility in Switzerland where LLW is treated [21]. This process enables both the processing of combustible materials and the melting of metallic parts, concrete and other solid matter. The same process can be used to vitrify organic and inorganic matter in the residue ready for final storage. All organic matter is totally decomposed.
Plasma treatment facilities are used widely in the treatment of hazardous wastes with some application in the nuclear waste processing field demonstrating that safety cases can be developed to support operation [22]. While plasma may be ideal for destruction of organics,the burden on the off-gas can be considerable and variable feedstock can challenge the operational stability on the plasma system particularly when using smaller crucibles.

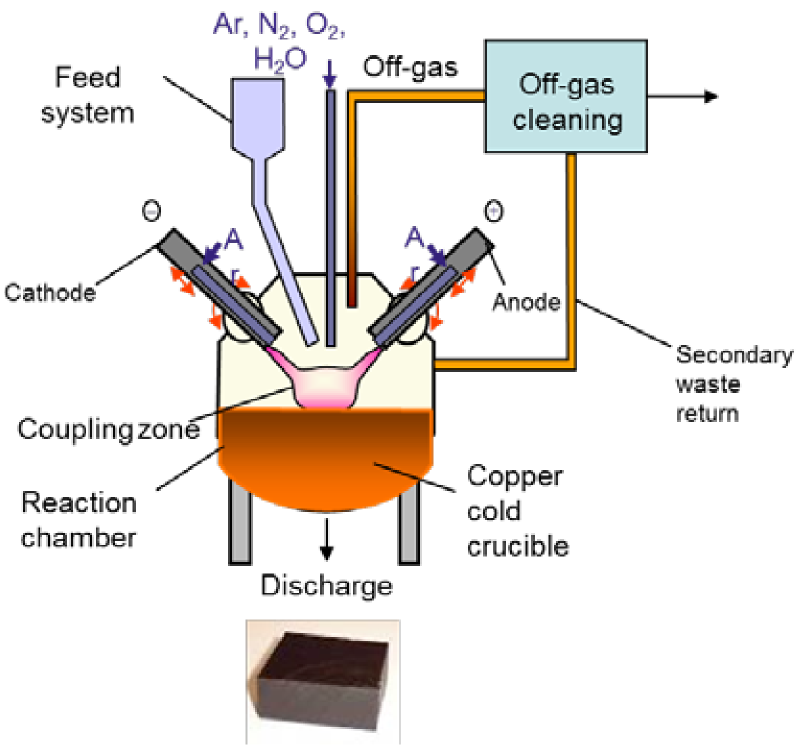

Figure 5: *Schematic of Plasma Treatment System*

Technology Options continued

Hot Isostatic Pressing

An alternative to traditional vitrification, Hot Isostatic Pressing (HIP) was originally adopted by the nuclear industry for the manufacture of fuel. It has been developed latterly for the immobilisation of Pu residues at Sellafield and of wastes from radioisotope production at ANSTO's Lucas Heights facility in Australia [23,24]. In a batch process HIP is used to consolidate pre-treated waste, premixed with the additives required, to produce a ceramic or glass-ceramic product. The simultaneous application of high temperature and pressure provides the conditions where a pre-ordained ceramic crystalline phase capable of hosting specific nuclides can develop. Typically wasteforms will be crystalline, ceramic often mimicking natural minerals which have been shown to remain unaltered over geological time periods. Glass ceramic composites can also be produced with the flexibility of the glass ceramic phase complementing the integrity of the ceramic.

An example of a glass ceramic composite wasteform is shown in figure 6 with a lathlike zirconolite crystalline phase used to immobilise uranium and plutonium surrounded by a flexible glass phase capable of immobilising other miscellaneous species.

While the consolidation process is relatively straight forward, pre-treatment steps are required to render the feed materials suitable for consolidation. This requires the removal of moisture and organics.

HIP is a batch process and while its use in the nuclear waste immobilisation field has been limited to kg scale, it is known to have been deployed in other industries at the tonne scale. As a batch process it is ideal for Pu and criticality management.

As the feed contents are totally independent of the energy input, it lends itself to processing of varied feeds such as orphan/niche wastes.

The development of glass ceramics has also shown how HIP can be used to treat heterogeneous wastes. The HIP itself can produce highly robust wasteforms. However, to take advantage of this capability the feed mix of precursor and wastes need to be in an appropriate form. Thus the challenges lie in the pretreatment stages and in defining formulations for HIP that reduce the potential loss of radionuclides at this stage.

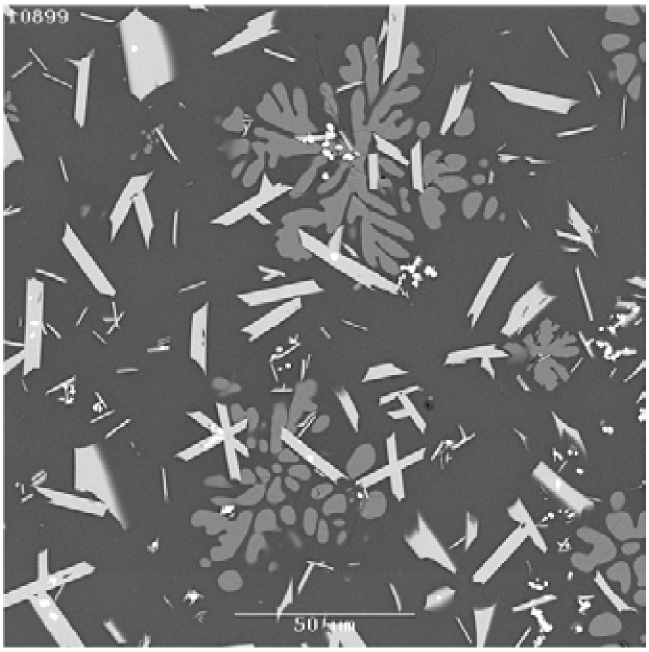

Figure 6: *SEM micrograph showing a glass ceramic composite wasteform* [25]

Metals melting

Decommissioning of nuclear facilities is resulting in metallic wastes. Routing such metals directly to a repository would not be best practice as it would take up valuable repository space and would be a waste of valuable resources.

A preferred alternative is to melt the metals first, in order to reduce the volume of waste. Using appropriate fluxes, metal can also be decontaminated - leading to the potential of re-use within the nuclear industry. The radionuclides which partition to the slag phase can then be disposed of. Data suggest that this thermal treatment option could prevent the unnecessary disposal of 95% of the volume of material. Typical examples are the operations carried out by Siempelkamp in Germany (26) and by Studsvik in Sweden. [27]

Smelting of metals is a well established technology. Its use in nuclear waste processing is largely to reduce the amount of material that requires geological disposal. Decontamination to below clearance levels may be achievable for lower active components that have some surface contamination, but this is unlikely to be achievable for metals containing a significant amount of activation product. Therefore the development challenge for smelting is to improve control of the chemistry and behaviour of melts to optimise removal of active components into a slag phase so the remaining metals can be shown to be suitable either for regulatory clearance or for re-use.

Hybrid systems

Thermal treatment methods can be combined to provide more complete and tailored solutions for specific waste streams. For example a facility could use a plasma system coupled with an inductively heated crucible system to provide optimum destruction of organics while tightly controlling melt characteristics to provide well conditioned, homogeneous void free products. The challenge here would be to avoid overcomplexity in ensuring multiple systems can work in concert efficiently.

Conclusions

Operational experience with UK HLW vitrification and international experience with a range of other wastes gives confidence that thermal treatment processes could provide feasible solutions to immobilise a range of UK wastes. These could include legacy wastes, decommissioning wastes and wastes arising from future fuel cycles.

Thermal processes offer potential benefits relative to the baseline technology of cement encapsulation, such as lower waste product volumes for storage, transport and disposal and improved compliance with key requirements of the waste package specifications, for example in the destruction of organics.

The development costs associated with bringing a single process and waste stream into operation and subsequently optimising the process are very substantial. A well-planned R&D and operational support programme can significantly reduce these costs, as was the case in the UK's Vitrification Test Rig (VTR) programme and its support to the vitrification of HLW.

A number of the technical challenges and development requirements to be addressed are common across the candidate thermal processes and wastes. For example radioelement volatility and off-gas treatment, process control and product testing. These common features offer the opportunity for cost efficiency in the comparison and development of thermal processes.

Overall development costs to bring a thermal treatment process into operation for UK ILW could be minimised and an effective comparison of candidate technologies could be made by addressing common R&D activities as a cross-industry programme. This would provide a test bed environment for multiple candidate processes by providing generic support capability such as active waste handling, off-gas treatment, process monitoring and product characterisation.

While specific design and choice of technology type will be a question for site licence companies, such early demonstration of candidate technologies in an active environment would help to inform interested parties and allow informed decision making.

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