

Demonstrating a Glass Ceramic route for the Immobilisation of Plutonium containing Wastes and Residues on the Sellafield Site

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ABSTRACT

A wide range of plutonium containing wastes and residues are currently stored on the Sellafield site. These wastes and residues arising from early fuel development activities at Sellafield are diverse in nature and in the absence of a long-term disposition strategy, ongoing storage represents a major financial commitment. An immobilisation technology is being developed, producing a glass ceramic wastefrom that will be suitable for extended periods of storage and ultimate disposal. Hot isostatic pressing [HIP] has been chosen as the preferred consolidation route. Following demonstration of the chemical feasibility of the approach at ANSTO's Lucas Heights facility, the project will demonstrate the technology by designing and building a full-scale inactive facility at Nexia Solutions' Workington site supported by a small scale active facility in the Technology Centre (BTC) at Sellafield. Delivery of this technology will allow the project sponsors, British Nuclear Group, the option of engineering a full scale plant in order to immobilise the actual waste arisings and discharge the liability to site.

INTRODUCTION

Previous processing of plutonium containing materials on the Sellafield site has resulted in a range of wastes and residues, from which it may be uneconomic to recover the plutonium. A quantity of these materials, weighing ca 0.5 Te, is of a diverse nature and as such will need to be treated on a case by case basis. At present these wastes continue to be stored and monitored and require periodic re-packaging, to prevent degradation of the packaging materials and possible release of plutonium contaminated material into the environment. Constant monitoring and the ongoing requirement for periodic re-packing are costly and any intervention inevitably involves dose uptake to operators. In their current form they present an ongoing liability to the newly established Nuclear Decommissioning Authority (NDA). A solution is therefore required to convert the wastes into a form suitable for passive safe storage and eventual repository disposal and thus removing the liability. In addition to the original 0.5 Te slated for immobilisation the possibility has been raised that the process could be used for additional materials on the Sellafield and other UK sites. In particular residues from the fabrication of MOx fuels, which

maybe uneconomic to feed back into the process, and plutonium oxide that has become heavily contaminated with chlorine as a result of long term storage in PVC bags.

Nexia Solutions, on behalf of British Nuclear Group in support of its plutonium management strategy, has been collaborating with the Australian Nuclear Science and Technology Organisation (ANSTO) to define technical options for the immobilisation of these residues. A batch process based on a glass-ceramic wasteform is proposed using a consolidation technology capable of accommodating the heterogeneity of the waste materials. Sufficient flexibility is being built into the process so that fully optimised process windows can be employed which will allow predictable product quality of the wasteform in a production environment. The proposed consolidation route must also be safe and suitable for use in a radioactive environment. Accordingly, a Hot Isostatic Press (HIP) process and associated glovebox line is being developed as a processing option that will enable the waste material to be successfully immobilised in a mechanically and chemically stable, and proliferation resistant, glass-ceramic or ceramic wasteform.

THE WASTES

The wastes and residues that were specifically targeted as feedstock for this particular process are contained in specially designed aluminium or stainless steel containers and are often double canned with additional PVC packaging in some cases. The residues are physically and chemically heterogeneous and have been categorised into different classes or families with the original aim being to identify specific treatment routes for each family. The plutonium content in these wastes is too low to make an economic proposition for plutonium extraction, but too high to allow them to be disposed of through a Plutonium Contaminated Material disposal (PCM) route. In fact levels of plutonium content are perhaps two orders of magnitude greater than the limits allowed under PCM disposal rules.

The majority of the waste consists of a number of families containing relatively small volumes of material and containing a mixture of both metals and oxides with a broad range of particle sizes requiring a flexible approach to treatment and immobilisation.

The actinide content of the wastes ranges from plutonium and uranium to smaller masses of neptunium and thorium. In some cases the plutonium is alloyed with elements such as aluminium and gallium.

The physical nature of the materials range from caked sludges, powders, lumps, pellets typically of around 1 cm in diameter and individual fuel pins. Cermets consisting of uranium and plutonium oxides on stainless steel plates are also included, the mixture of ceramic and metal presenting a particular challenge to the treatment process. A wide range of minor non-active components, including organics, is also reported to be present in various residue families. Overall the plutonium content is expected to be no greater than 50 kg with the total weight of the wastes being ca 0.5te.

The development of such a process will also allow options for the treatment of other plutonium containing materials which may require an immobilisation option dependent on the economics of recovering the plutonium and on the need for plutonium as a feed material for future fuel. These wastes include plutonium dioxide which has been heavily contaminated by chlorine as result of storage for long periods of time in PVC bags and residues produced during the development and production of MOx fuel which may be deemed unsuitable to feed back into the process.

While the wastes remain safely stored in the short term there is a necessity to demonstrate a process suitable for creating a wasteform acceptable for long term safe storage at Sellafield prior to disposal in an as yet undefined repository.

THE TECHNOLOGY

The solution proposed for the immobilisation of these wastes includes the use of a glass-ceramic wasteform providing the advantage of combining the necessary processing flexibility of glasses with the chemical durability of ceramics, which are most suited for immobilising actinides. The key crystalline phase in the glass-ceramic is zirconolite (ideally $\text{CaZrTi}_2\text{O}_7$), a naturally occurring titanate mineral and the main host for plutonium and other actinides in Synroc. Zirconolite has considerable chemical flexibility, with extensive ionic substitutions in its structure possible involving a large number of other cations in addition to the actinides [1]. The scientific basis for its use in the immobilisation of actinides has been extensively studied and peer-reviewed [2, 3, 4, 5, 6]. Furthermore, extensive complementary studies on naturally occurring zirconolites have confirmed that they survive in the natural environment and are capable of locking up actinides in the crystalline matrix for millions of years [2, 7, 8, 9]. Zirconolite is not only extremely chemically resistant but it can also be synthesised throughout a wide range of waste form chemistries [6]. The vitreous phase of the glass ceramic will be used to accommodate the widely varied non-actinide components of the waste stream.

The required minimisation of aerial and liquid effluents determined that a "dry" process be employed. It was felt that the chosen wasteform could be produced through either traditional ceramics routes of forming and consolidation or through a melting route. A thorough review of processing technologies was carried out [10] in order to determine the appropriate consolidation and pre-treatment options required to produce the glass ceramic wasteform. Hot Isostatic Pressing was chosen as the consolidation mechanism and when integrated with the glass-ceramic waste form, the HIP consolidation step contributes to a highly flexible process with the ability to accommodate the large variation anticipated in the waste feed streams under conditions that promote reaction of refractory waste components such as PuO_2 and create a highly uniform product.

Pre-treatment of the wastes is required prior to acceptance for the Hot Isostatic Pressing Process (HIP). The ideal feed for the HIP is a fully oxidised material with an appropriate particle size consistent with the creation of a glass ceramic. The varied nature of the wastes dictates that a combination of pre-treatment technologies is required. Firstly the varied physical nature of the wastes needs to be treated by a series of size reduction steps. Work to date [10] has identified the need for two or three pieces of equipment capable of reducing to feed to appropriate size. Jaw crushing and disc mills will be used to reduce material from anything up to 100mm in diameter down to the level required to make a good glass ceramic. Secondly there is a metallic component to the waste feed and a rotary calcination step is being proposed having the option to use either ceramic or metal tubes, the former being necessary should oxidation temperatures of $>1000\text{C}$ be required. Once the materials have been fed through the calcination and size reduction steps, they are blended with appropriate precursors and subsequently granulated in order to provide the feed to the HIP. The basic outline of the whole process is shown in Fig. 1.

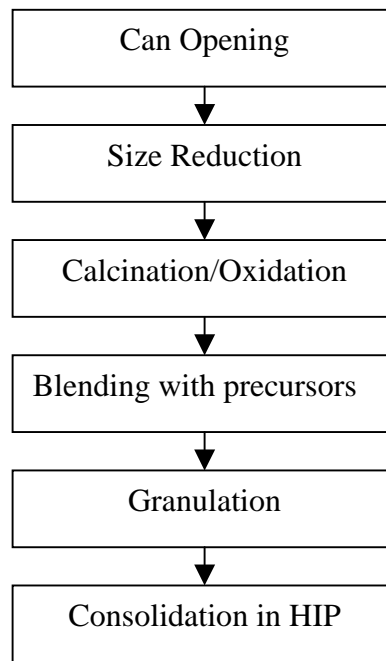


Fig. 1. Outline of the Hot Isostatic Pressing Process

DEVELOPMENT

Feasibility

The challenge to the immobilisation of the named wastes and residues mainly comes from their diverse chemical and physical nature. With appropriate formulation design, combined with the HIP process, the Pu and other actinides in the residues can be directed to partition overwhelmingly into zirconolite at the expense of the vitreous phase. The vitreous phase immobilises the glass forming constituents and provides the chemical flexibility necessary for the highly variable waste streams envisaged.

The feasibility of the approach has been demonstrated by obtaining baseline data validating the optimum processing conditions for the Pu bearing glass-ceramic waste form. Different zirconolite glass-ceramic formulations were tested in order to accommodate a span of possible waste loadings. These formulations were tested initially with U and Th as surrogates and then subsequently validated with Pu. The most fundamental aspects of waste form properties that were addressed in the demonstration were the effects of in-can redox, temperature, and Pu concentration (waste loading) on:

- Phase development and Pu partitioning (also comparing the behaviour of Pu with U and Th surrogates)
- Aqueous durability of the waste form, especially with respect to Pu and an added Gd neutron absorber
- Resistance to retrieval of the Pu from the waste form upon simple crushing and dissolution in concentrated common mineral acids such as H₂SO₄ and HNO₃.

The intention has been to scope out a wide processing window; for instance a five orders of magnitude wide range in oxygen fugacity [from the FeO-Fe buffer to NiO-Ni], 100° C wide temperature range, and a range of waste loading from 20 to 40%.

A demonstration was carried out on the basis of a model blended waste stream composition. This composition was estimated from historical data by taking into account the major residue families and determining a weighted average composition for the major constituents, based on the relative amounts of material in the major families.

Uranium, initially as metallic swarf, and later as U_3O_8 , was used as a surrogate for metallic or completely oxidised original Pu metal respectively. ThO_2 (and also CeO_2 initially) was used as a surrogate for any potentially coarse grained refractory PuO_2 in the wastes. U and Th were both used as simulants for Pu to maintain consistency with the process options evaluation that has progressed in parallel with this work. For example, refractory high-fired ThO_2 is being used as a simulant for any possibly refractory PuO_2 in the size reduction optimisation trials. In addition, it was decided that two actinides acting as simulants for the ranges of Pu behaviour possible under a range of experimental conditions are a more rigorous evaluation than a single actinide (or a single lanthanide such as Ce).

The experimental programme for demonstration of the feasibility is described fully elsewhere. [11,12] Wasteforms made with U + Th were prepared on a scale of around 250 g and validated using plutonium at a 35 g scale. In addition U + Th containing samples were made at the 35 g scale to demonstrate that wasteform properties were independent of scale.

The waste simulants were made by mixing SiO_2 and CaF_2 , representing a slag component present in some waste feeds, with either U + Th or Pu. This was then mixed with the wasteform precursor, formulated to produce zirconolite crystals in a glass matrix. The materials was HIPped at a range of temperatures from 1220C to 1320C.

After HIPping the samples were characterised for overall structural integrity, phase development and crystallinity, phase composition and actinide partitioning, via optical microscopy, scanning electron microscopy coupled with energy dispersive spectroscopy (SEM/EDS) and x-ray diffraction (XRD).

Aqueous durability was evaluated using a modified replacement MCC-1 leach test with deionised water as the leachant. Proliferation resistance was demonstrated by treating 2 g of crushed wasteform in the 75-150 μm particle size range with 5 M nitric or sulphuric acid for one week at 22° C.

A typical wasteform microstructure is shown in Fig. 2. As intended, zirconolite and calcium fluoride are abundant as revealed by both XRD and SEM/EDS. In addition, minor zirconia is present and metallic Fe or Ni, depending on the metallic buffer used. This indicates that the intended redox conditions have prevailed throughout the HIP cycle. The light phase shown in the figure is zirconolite, the grey phase is CaF_2 and dark matrix is the vitreous phase.

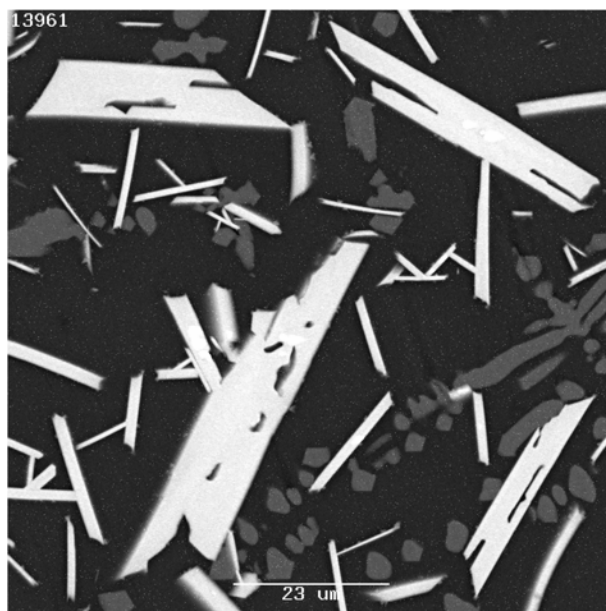


Fig. 2. SEM micrograph of glass ceramic

Actinide partitioning is a function of actinide type, waste loading and oxygen fugacity during HIPping. For the Ni metal buffered runs the partitioning coefficient of Pu and U - defined as wt% Pu in zirconolite divided by wt% Pu in glass - was of the order of 100; typically the U or Pu in the glass was below the detection limit which was then used as a notional U or Pu content for the glass to calculate the partitioning coefficient.

The partitioning of Pu into zirconolite was less strong for the 24 wt% waste loading sample HIPped with an Fe buffer. In this case the coefficient was 20 and this could be due to an increased level of Pu³⁺ which will be more easily incorporated in the glass matrix.

Leach data are reported comprehensively elsewhere. [11] Pu leach rates over a 28-day period were in the range 10^{-5} to 10^{-4} gm⁻²d⁻¹ and are consistent with actinide leach rates from fully crystalline titanate ceramics. Analysis of leachates for Gd, U and Th were typically below detection limits, and leach rates, calculated from the detection limits, were of the same order of magnitude as those for Pu.

The concentrations of Pu dissolved by the mineral acids in the plutonium retrievability tests were used to calculate the number of kilograms of wastefrom that would need to be diverted, crushed and leached in order to access 1 kg of Pu. For optimum fabrication conditions, the mass of wastefrom that would need to be diverted would be in excess of 10,000 kg.

Further development is being carried out to demonstrate flexibility and an approach that would widen the envelope to immobilise the variation in the feed and at the same time reducing the complexity of operation.

Actinide oxidation

This initial experimentation has demonstrated that a glass ceramic, or indeed in the case where the feed has minimal impurity levels, a full ceramic wastefrom can be produced. In order to prepare a suitable feed for the production of a ceramic or glass ceramic it is necessary to fully oxidise the feed material. An investigation was therefore carried out into oxidation of plutonium and uranium alloys, simulating some of the feedstock that it was felt may be most problematic to oxidise. The feedstock for the investigation included plutonium metal, aluminium encapsulated uranium silicide and aluminium encapsulated uranium fuel. These were felt to represent the variety of material types found in the residues feedstock on the Sellafield site.

Oxidation temperatures for plutonium of 400C and 600C were explored in air. Only limited oxidation was observed at the higher of the temperatures. The atmosphere was then changed to moist air and moist nitrogen again at the temperatures of 400C and 600C. Complete oxidation was found in moist air or nitrogen at 400C. The ability to calcine at the lower temperature in moist nitrogen significantly reduces the pyrophoric risks associated with the process.

In contrast to the ease in oxidising plutonium, difficulty in oxidising uranium has been encountered in treating aluminium encapsulated uranium metal or disilicide fuel and shows that for economic treatment of like materials it may be necessary to find an alternative treatment route.

Metal Encapsulation

Metal Encapsulation has been shown to provide a route to the small minority of waste where investment in time and equipment necessary to produce glass ceramic wastefroms would not be either cost effective or justifiable in terms of operator dose and increased effluent. The route utilises the same HIP as for the ceramic and glass ceramic wastefrom, but the feed is left largely intact and surrounded with copper powder prior to HIPing. The philosophy is akin to that employed by Sweden in their concept for spent fuel. Preliminary tests on fragments of aluminium uranium dispersion fuel, graphite-carbide and CERMET parts show that after consolidation at a relatively low temperature of 360C, the parts remained stable and encased within a copper matrix. This approach has not yet been optimised, but these early results show promise in using this technique for immobilising such minority feeds.

Oxidation of carbides and graphite

Carbides and graphite are known to form part of the stock of residues under consideration. The process as designed would entail the oxidation of these components prior to adding precursor and then feeding into the HIP can for consolidation. The degree of difficulty involved in oxidation of high levels of certain species may dictate that residual levels of carbide and graphite remain after calcination and therefore an assessment was made as to the level that could be tolerated in both ceramic and glass ceramic wastefroms.

Calciner trails suggest that at 1000C graphite would be removed from the wastestream although carbides in this case in the form of silicon carbide would remain behind. Early studies suggest that at levels of ca 1% remaining silicon carbide or carbon have little effect on the formation of the zirconolite in the glass ceramic and the subsequent partitioning of the plutonium simulant to the crystal phase. In the case of full ceramic residual carbon and silicon carbide can lead to

increased porosity but no or little apparent effect on phase formation and thus wastefrom performance.

Chlorine

The future waste inventory may comprise of materials containing significant levels of chlorine resulting from storage of plutonium oxide powders in PVC bags. Chlorine levels up to 0.1 wt% can be expected. While the calcination step has as its function the elimination of halides and other such materials from the feed in preparation for the HIP, it is possible that some chloride may remain in the material to be fed to the HIP. As such it has been important to demonstrate that these levels have zero or negligible effect on either the process or the resulting product.

To that end full ceramic samples, both pyrochlore and zirconolite, have been produced at levels of chloride of up to 0.2% using the HIP technology. No detrimental effect on the nature of the ceramic and the partitioning of the plutonium to the zirconolite or pyrochlore phases was exhibited and neither was there any sign of attack on the HIP can from the chlorine during the consolidation process.

Process equipment optimisation

A key stage of process development has been to demonstrate the particle size that can be tolerated whilst still producing a wastefrom meeting the required performance specifications. Knowledge of this tolerance is critical in making decisions regarding the design of a full-scale process line.

The necessity for size reduction has been determined from a physical inventory of the feedstock. Although the line is being specifically designed for the production of a glass ceramic wastefrom, the need has also been identified for the production of full ceramic wastefroms where appropriate as these may reduce overall wastefrom volumes.

In the case of the glass ceramic wastefroms, liquid phase sintering may play a part in the consolidation step. With the level of liquid phase present, it is possible that the level of particle size reduction may be reduced. The ability to use larger particle sizes may have beneficial consequences for any active plant from potential contamination and effluent handling problems that may arise with fine powders as well as the elimination of a fine grinding process step.

Recent experimentation has been carried out comparing the particle size requirements in producing high quality glass ceramic and ceramic wastefroms HIPed at temperatures of between 1270C and 1320C. At the higher temperatures, the process was shown to be capable of producing a glass ceramic when fed with particles of between 500 and 800 microns. In the case of a full ceramic formulation, particle size reduction of down to 20 microns was found to be necessary.

These results demonstrate the possibility of limiting the size reduction steps in the case of glass ceramics with the concomitant reduction in effluent production. Although finer size reduction is required in the case of ceramic wastefroms, it is likely that in these cases that size reduction step will be the only one required.

DECISION MAKING

An analysis of the families and knowledge of the capability of the wastefrom has allowed the waste to be allocated to three classes of wastefrom. The overall design philosophy for the process line is to direct the waste into the flexible glass ceramic matrix, unless there is a clear driver to do other wise.

- Glass ceramic - which will be used for the majority of families due to the flexibility of the vitreous phase in accommodating the varied feedstock.
- Full ceramic - to be used in cases where the residues are relatively pure oxides of uranium and plutonium and the use of a full ceramic composition would reduce the overall level of waste containers produced. Although covering fewer families these may constitute the higher volumes feed.
- Metal encapsulation - to be used in a small minority of cases where it may be difficult to oxidise all the feed components required for the formation of a ceramic or glass ceramic.

The decision trees shown below in Fig. 3. and Fig. 4. indicate the key factors and decision points that influence the selection of a given immobilisation matrix.

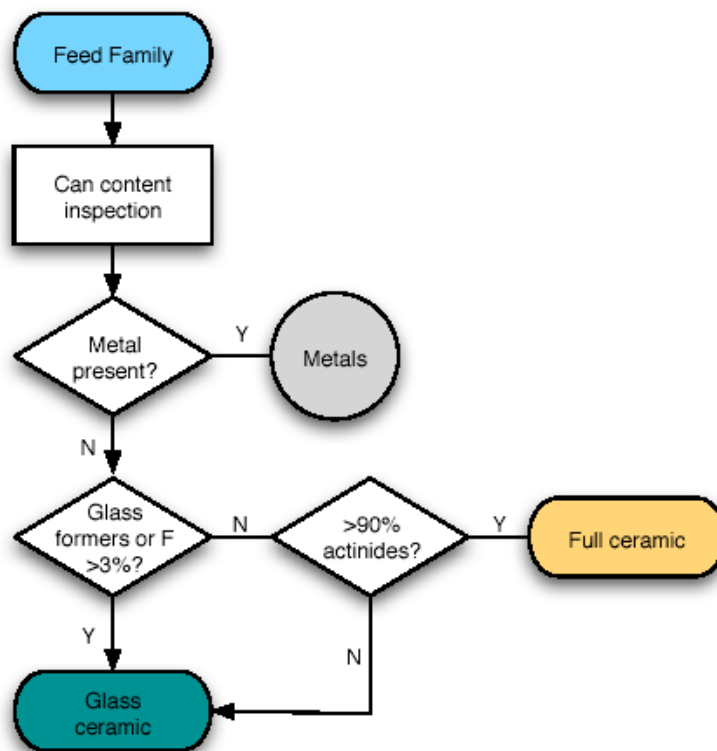


Fig. 3. Decision tree for waste families

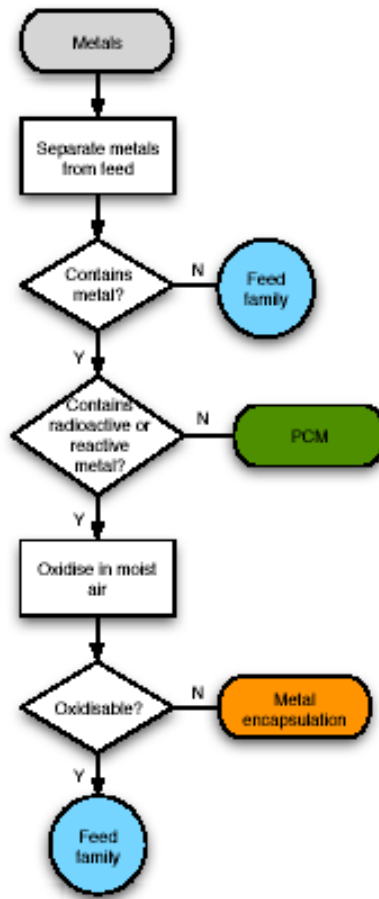


Fig. 4. Specific decision tree for high metal wastes

WASTEFORM ACCEPTANCE

Wasteform development to date has shown the capability of this technology and has allowed us to set draft parameters for wasteform acceptance. Any plutonium containing material produced through this process will need to comply with conditions for acceptance to the new Sellafield Product and Residue Store (SPRS). Both handling and criticality constraints currently limit the waste package size to around 20kgs. The process is being designed in order that it can produce a package of that size in a single HIP cycle.

While specific packaging for the repository will need to be determined when repository concepts have been finally been agreed upon, the project aims to develop wasteform parameters that will enable the wasteform package to be disposed of in a range of repository concepts.

The development programme aims to show that for a wide range of feeds a product can be delivered which demonstrate an appropriate level of durability measured against the possible environment in which it may be disposed. In the case of the glass ceramic, partitioning of plutonium to the crystalline phase can be a measure of that durability and process conditions will be developed in order to achieve that partitioning. It is important to note that in the final process, quality will be determined by demonstrating process control thus negating the requirement for routine testing of the final product.

Proliferation of nuclear material is an area of key concern. Maximisation of partitioning of the plutonium into the more durable ceramic phase is clearly of benefit and a measure of the intrinsic retrievability has been determined by attempting dissolution with mineral acids [11].

The parameters of durability and retrievability can be affected by radiation damage. As such a package of work has been initiated which will the glass ceramic product will be subject to radiation damage by the incorporation of Pu-238. The performance of the material will be assessed a period of time its ability to withstand determined.

DEMONSTRATORS

In order to demonstrate the capability of this process a series of waste variation studies will be carried out. Major precursor components, impurity cations and levels of anion impurities will be assessed in a matrix of experiments. This will enable the appropriate recipes to be compiled for the required class of wasteform.

Once recipes have been determined process operation will need to be fully demonstrated. Only after this is done can the work commence on nuclearisation. The method to be used is two fold. A full scale inactive facility is being designed and will be constructed at the Nexia Solutions facility in Workington, Cumbria. This facility will mimic all aspects of the process and will deviate from the envisaged process only in the use of cerium as a simulant for uranium and plutonium. This facility will be used to define and optimise all of the operating parameters of the final active plant. Because key aspects of the process involve size reduction of the residues and ensuring homogeneous blending of residues and wasteform precursors, refining of the processes and parameters can be conducted without any actinide species being involved.

However it is recognised that the redox and partitioning behaviour of cerium will not accurately replicate that of the actinides, and accordingly a small-scale active glovebox facility is being built on the Sellafield site. This will allow wasteform development work to be carried out for which the use of plutonium and other actinides is essential. It will also allow for the validation of data obtained from the Workington facility particularly relating to wasteform chemistry. The design and safety work essential to enable the construction of such a facility will itself address any issues that there may be over the operation of such a process on a UK nuclear licensed site. The aim is that the HIP itself is operated outside the glovebox environment, being in itself a form of containment. Preparation of material for the HIP, this will include a calcination/oxidation facility, will be carried out in 4 glove boxes with provision to maintain the can outside the glovebox during the filling procedure. This facility will also enable simulated wasteforms to be examined by standard materials characterisation techniques and subjected to leaching and plutonium retrievability trials.

CONCLUSION

A glass ceramic wasteform and process is being developed that has the potential to immobilise a range of plutonium containing residues stored at the Sellafield site that have diverse compositions and physical characteristics. The wasteform demonstrates excellent leach resistance for plutonium. The processing route avoids the use of water and other liquids, thus minimising the generation of secondary effluents. A metal encapsulation technique is being developed in minority cases where the production of a glass ceramic is not cost effective.

The process will also be capable of immobilising other possible future waste arisings including chloride-contaminated materials and MOx residues in a full ceramic wasteform.

The process will be demonstrated at full scale inactively, followed by small validation using plutonium prior to transfer to an operating unit for design and build of a full-scale plant capable of discharging the liability.

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