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The HADES Facility for High Activity Decommissioning Engineering & Science: part of the UK National Nuclear User Facility

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Abstract. Research and innovation is key to delivering UK Government's civil nuclear energy policy, in particular to accelerate reduction in the hazard, timescale and cost of legacy decommissioning and geological disposal of radioactive wastes. To address this challenge, a national centre of excellence, the HADES Facility, has been established to support research and innovation in High Activity Decommissioning Engineering & Science, as part of the wider network of UK National Nuclear User Facilities. Herein, we describe the development of this user facility, the current status of its capability, and functional equipment specifications. The unique capabilities of the HADES Facility, in the UK academic landscape, are emphasised, including: handling of weighable quantities of ^{99}Tc and transuranics; quantitative electron probe microanalysis of radioactive materials; hot isostatic pressing of radioactive materials; and laboratory-based X-ray absorption and emission spectroscopy. An example case study of the application of the HADES capability is described, involving thermal treatment of a real radioactive ion exchange resin waste to produce a conceptual vitrified waste form.

1. Introduction

The UK Government's civil nuclear policy has the fourfold objectives of: exploiting nuclear power for low carbon, reliable and affordable electricity generation; decommissioning of legacy nuclear fuel cycle activities; clean-up of defence nuclear liabilities; and safe disposal of radioactive wastes [1-7]. Building on the Nuclear Industrial Strategy, the Nuclear Sector Deal set the vision for the civil nuclear sector, to deliver by 2030 [3]:

- 30% cost reduction in cost of new build projects.
- Savings of 20% in cost of decommissioning compared with current estimates.
- Up to £2 billion domestic and international contract wins.

Research and innovation is recognised as crucial to meeting these challenges. In particular, there is a need for disruptive technologies to deliver safe and cost-effective management and disposal of radioactive wastes from legacy and future advanced fuel cycle operations. At the forefront of such technological development is thermal treatment of radioactive wastes, which utilises the application of heat to immobilise radionuclides and chemotoxic species within a passively safe product (a glass, slag or ceramic) suitable for interim storage and disposal. The benefits afforded by this approach include volume reduction (by removal of water, volatiles and voidage) and elimination of waste reactivity and organic inventory (by oxidation or thermal destruction). Nevertheless, technology maturation and implementation are critically dependent on [1, 8-10]: development of wasteform formulation and process envelopes; control of radionuclide volatility; understanding of product heterogeneity; and evaluation of wasteform behaviour in a Geological Disposal Facility (GDF). With regard to the latter, the Nuclear Decommissioning Authority (NDA) and its subsidiary, Radioactive Waste Management Ltd. (RWM), highlighted the need for research to understand the disposability of thermal treatment products, including those for plutonium disposition [1, 7-10].

NDA Strategy 3 prioritised the need to develop higher activity waste strategy in a number of areas where there is the potential for beneficial change [1], recognising that cement encapsulation was unlikely to be the optimal solution for all future waste streams. Accordingly, the new NDA 5-year R&D Plan 2019-24, prioritises as a Key R&D Topic – Alternative waste treatment, with a particular focus on volume reduction understanding and, where appropriate, addressing the technical barriers to *implementation of new thermal… treatment technologies across the NDA group* [10]. From a financial perspective, the potential reduction of 50% in packaged higher activity waste volume achieved by thermal treatment, could translate into UK-wide cost savings in intermediate level waste (ILW) packaging and storage costs, potentially of up to £1 billion [8].

Targeted estate-wide deployment of thermal treatment technologies could play a pivotal role in realising the 20% reduction in decommissioning costs demanded by HM Government’s Nuclear Sector Deal [3]. The need for such intervention was recently highlighted by the House of Commons Public Accounts Committee [11, 12], who recommended NDA focus on delivering value for money to tax payers in decommissioning the Sellafield site and accelerating plutonium management strategy. To progress deployment of thermal treatment technology in UK decommissioning, NNL have invested in an active Geomelt^{TM} thermal treatment demonstrator on the Sellafield site, with inactive alternate technology demonstrator systems developed by the commercial sector, such as the Tetronics – Costain partnership to develop plasma vitrification technology. Whilst these pilot scale demonstrators have proven effective in building confidence in thermal processes and products, they rely on upstream small-scale laboratory research to design, develop and initially constrain formulation and process envelopes, and to gain early insight into downstream challenges such as off gas burden and product disposability. The window for insertion of thermal treatment technologies into lifecycle baseline plans is 2022-40 [8-9], and hence investment in enabling research and infrastructure is exceedingly timely.

We identified a national need for state of the art laboratory research infrastructure and equipment to ensure timely deployment of thermal treatment technology to meet the objectives of NDA Strategy and the Nuclear Sector Deal. To address this strategic need, the Engineering & Physical Science Research Council (EPSRC) and The University of Sheffield, announced, in spring 2020, the investment of £1M to establish the HADES Facility for High Activity Decommissioning Engineering & Science at The University of Sheffield, as part of the National Nuclear User Facility network. This leveraged prior recent investment of ca. £8M in existing state of the art infrastructure for radiological materials science, from UK Government, The University of Sheffield, EPSRC, NDA, and the Royal Academy of Engineering, and others.

2. Description of the HADES Facility

The HADES Facility is accommodated within ca. 400 m^2 of state of the art radiochemistry laboratories at The University of Sheffield. All laboratories are designated as Supervised Areas with relatively low hazard work with radionuclides permissible throughout the facility (kBq nat. U/Th). Additionally, there are Controlled Areas for more hazardous work with low inventories of radionuclides with high specific activity (100 kBq nat. U/Th; MBq ^{99}Tc; 10 kBq other β,γ). Investment of £1M by EPSRC and the University of Sheffield will upgrade a recently refurbished radiochemistry laboratory (60 m^2) to a Restricted Controlled Area, to satisfy demand for research using substantial inventories of radioactive materials, e.g. in a single procedure: 10 MBq of natural U/Th, or 5 MBq of mixed β,γ nuclides (e.g. ^{3}H, ^{14}C, ^{60}Co, ^{90}Sr, ^{137}Cs). Importantly, this will grow UK academic capacity and capability to meet demand for working with small quantities of real radioactive wastes, such as recent research on thermal treatment of spent ion exchange resins with an activity of 10 MBq kg^{-1} β,γ, contact dose rate 150 μSvh^{-1} – see case study in Section 4. In 2020, we will also commission a Restricted Controlled Area laboratory for working with transuranics within an integrated transuranic-materials glove box and furnace suite and a radiological Electron Probe Micro-Analyser (EPMA). These developments, funded by EPSRC as part of the Sheffield Hub of the Royce Institute, will deliver the first capability in the UK academic sector for working with weighable quantities of transuranic materials and accurate spatially-resolved microchemical analysis of radiological materials

3. HADES Facility Capability Platforms

Equipment and instrumentation within the HADES Facility is organised within the following capability platforms:

3.1. Materials handling

Negative pressure glove box workstations, dedicated for separate handling of U/Th and ^{99}Tc, are provided for safe manipulation of radiological materials under inert conditions (<ppm O_2, <ppm H_2O). For small radioactive inventories, which do not require handling under inert conditions, a suite of standalone containment glove boxes enable material manipulations under air or inert gas. Additionally, a standalone platform of three glove box workstations, with an integrated high temperature furnace (<1600°C) is dedicated to handling of transuranic materials.

3.2. Synthesis and processing

A suite of high temperature furnaces is available for the fabrication of glass, glass-ceramic and ceramic wasteforms, and investigation of thermal treatment processes, operating up to 1800°C under controlled atmosphere. Glass melts may be agitated by stirring or gas sparging, if required. Off gas analysis can be undertaken in real time by mass spectrometry or by chemical and radiochemical analysis of scrubber solutions. The HADES Facility also accommodates a unique capability in the UK for Hot Isostatic Pressing (HIP) of radiological materials using an AIP 6-30 HIP system with a custom built Active Furnace Isolation Chamber (AFIC); non-active and active materials may be processed up to 2000°C and 1350°C, respectively, under up to 200 MPa Ar. A suite of powder mixing, milling and grinding equipment is also available for high throughput batch preparation. A selection of non-stirred and stirred Parr pressure vessels are offered for hydrothermal and solvothermal syntheses.

3.3. Diffraction and spectroscopy

Rapid routine powder X-ray diffraction (XRD) analysis is undertaken using a Bruker D2 Phaser XRD system, with Ni filtered Cu $K\alpha$ radiation and a Lynxeye energy discriminating detector. A Panalytical X'pert 3 XRD system, with a parabolic mirror for parallel beam Cu $K\alpha$ radiation and PIXCel 1D silicon strip detector, is also available for high throughput data acquisition, grazing angle analysis, and in situ high temperature studies using an Anton Parr HTK 1200N furnace (up to 1200°C, under controlled atmosphere). Additionally, HADES is host to the UK's first laboratory end station for X-ray Absorption and Emission Spectroscopy (XAS / XES), which enables element-specific speciation studies in the range 4.5 – 18.0 keV, based on a customised EasyXAFS XES100 spectrometer, with ca. 1 eV instrument resolution achievable in the XANES region [13]. This unique capability is complemented by a Mossbauer spectrometer (Wissel MRG-500), for investigation of Fe and Sn speciation and local environment, including conversion electron detection for surface sensitive Mossbauer experiments (<100 nm for Fe). Confocal Raman spectroscopy with spatial and depth resolution, in the range 50-3200 cm^{-1} with 1.4 cm^{-1} spectral resolution, is enabled by an Horiba XPlora Plus system equipped with a 532 nm laser.

3.4. Microscopy and microanalysis

Imaging of microstructure and micro-chemical analysis is undertaken using a workhorse Hitachi TM3030 scanning electron microscope (SEM), with magnification up to x30,000 in back scattered electron mode, equipped with an integrated Bruker Quantax 75 Energy Dispersive Spectrometer (EDS). The Hitachi TM3030 is also capable of imaging in low vacuum mode, for imaging of non-conductive, environmental or samples in their natural state. Access to a Philips XL30 SEM with an Oxford Instruments INCA EDX system is available for analysis of U/Th materials which require high magnification and/or topographical analysis in secondary electron mode. In 2020, as part of investment through the Sheffield Hub of the Royce Institute, a new JEOL JXA-8530F Electron Probe Micro Analysis system will be commissioned to enable quantitative microchemical analysis of radiological and nuclear materials by wavelength dispersive spectroscopy (WDS). 3D surface characterisation and imaging of materials is also possible using a Bruker Contour Elite K profilometer and Bruker MultiMode 8HR Atomic Force Microscope (AFM).

3.5. Thermal and physicochemical analysis

A Netzsch TG 449 F3 Jupiter simultaneous thermal analyser enables thermogravimetric and differential thermal analysis or scanning calorimetry (TGA and DTA / DSC) of materials, with coupled mass spectroscopy of evolved gases (up to 1500ºC). For high throughput TGA-DSC analysis, a TA Instruments SDT 650 system is available (up to 1500ºC). Additional physicochemical characterisation techniques include: glass rheology (Theta Rheotronic II; 5 x 10^{-3} to 5 x 10^{-6} Pa s, up to 1650°C); surface area analysis (Beckman Coulter SA-3100); helium pycnometry (Micromeritics Accupyc II); and particle size analysis (Malvern Mastersizer 3000).

3.6. Chemical and radiochemical analysis

A Thermo Fisher Inductively Coupled Plasma Optical Emission Spectrometer and Mass Spectrometer (ICP-OES 6000 iCAP and ICP-MS iCAP RQ) are available for trace and ultra-trace analysis of aqueous solutions, depending on analyte and concentration. Both systems are equipped with auto-samplers, and the ICP-MS system is equipped with an auto-dilution module, to enable rapid sample throughput. Two Thermo Fisher Ion Chromotography systems (ICS 1100) enable dissolved cation and anion analysis, with electrolytic suppression for low noise and drift, reduced background and lower detection limits; a coupled auto-sampler is available for high throughput analysis. These capabilities are complemented by a Hidex 300SL liquid scintillation counter using Triple to Double Coincidence Ratio (TDCR) technology for absolute activity measurement and separation of α and β scintillation. Accurate bulk chemical composition is determined using a Panalytical Zetium wavelength dispersive X-ray Fluoresence Spectrometer, equipped with a 4kW rhodium X-ray tube for high count rate and sample throughput and large area mapping capability (300 μm spot size); this was the first such state of the art system installed in the UK. In the next phase of Facility development, additional analytical capability will be procured including analysers for total organic carbon and oxygen / nitrogen determination.

3.7. Wasteform alteration and dissolution

A suite of temperature controlled ovens are dedicated to short and long term alteration studies of wasteform alteration behaviour using standardised and tailored protocols. Both batch and dynamic experiments are possible, including single pass flow through methodology for accurate determination of kinetic dissolution parameters. If required, experiments can also be performed under anoxic conditions, including periodic sampling.

3.8. Radiometrics and radiological protection

A Canberra BEGe high resolution gamma spectrometry system is available for identification and quantification of radionuclides in solid or liquid samples. In controlled areas, fixed personal contamination monitors are available, supplemented by a suite of portable large area survey meters, contamination monitors and dose rate detectors, and an Identifinder R100 for initial survey of received packages.

4. Case study of thermal treatment research enabled by the HADES Facility

In the UK context, spent ion exchange resins (SIERs) are classified as both intermediate and low level waste; some resins are considered to be problematic *radioactive waste… which has no defined waste* treatment and disposal route available or for which existing routes are suboptimal [14]. Challenges with SIERs include: incorporation of complexants, such as EDTA (ethylenediaminetetraacetic acid), and degradation of material in disposal environments to yield chelate molecules, which facilitate rapid transport of complexed radionuclides; and degradation of material during storage to yield chemically toxic and volatile benzene, phenol, trietheylamine, and potentially ammonia gas [15-17].

One approach for effective conditioning of SIERs is thermal treatment utilising suitable glass forming additives to yield a passively safe glass or slag (crystallised glass) type wasteform [18-21]. We have investigated application of such an approach to support the strategic waste management considerations of a UK waste producer in partnership with Costain. In the present investigation the SIER waste is a mixture of Purolite NRW-100 strong acid cation and NRW-400 strong base anion resins, in a 1:1 chemical equivalent, with an admixture of EDTA complexant, see Figure 1. A full account of the research to demonstrate effective thermal treatment of this waste, at laboratory scale, will be published in due course. Here, we highlight some key outcomes of the second phase of the research programme, using real SIER waste, enabled by the capability of the HADES facility.

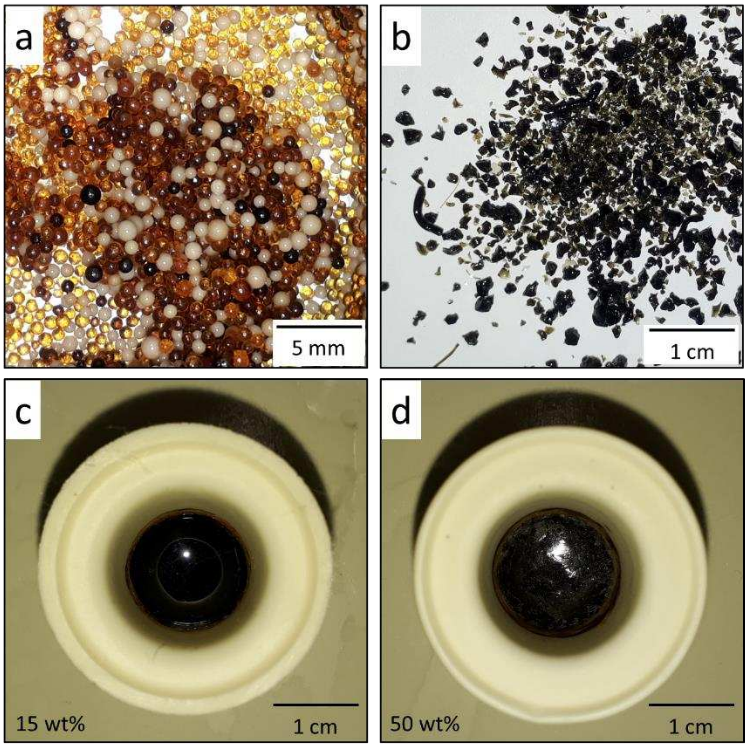

Figure 1/ a) Real spent ion exchange resin material; **b)** G73 glass frit; vitrified products produced from thermal treatment of **c)** 15wt% and **d)** 50wt% real spent ion exchange resin (on de-watered basis).

Table 1. Composition of G73 glass.

Component	Weight %
SiO_2	41.06
B_2O_3	2.06
Al_2O_3	0.28
Fe_2O_3	6.15
CaO	8.05
BaO	37.01
Li_2O	3.08

Our previous work had established the potential of barium borosilicate glasses for thermal treatment of SIERs, such as the G73 formulation summarised in Table 1, which has a high tolerance to sulphate formed by degradation of the resin sulphonate functionality [19-22]. Thermal treatment experiments at 10 – 40g scale were undertaken by first drying the resin to constant weight at 90°C and mixing with the required quantity of G73 glass frit in an alumina crucible, the mixture was ramped to 1000-1100°C at 3°C / min for 8h.

The first stage of our research showed that successful thermal treatment of inactive surrogate ion exchange resin at 1100°C, to yield a glass or slag product, could be achieved at an incorporation rate up to 50wt% of de-watered resin (20 wt% based on the dried resin equivalent), without separation of a sulphate salt. The real SIER material supplied for the purpose of research was expected to incorporate several inorganic radionuclides, e.g. ^{54}Mn, ^{55}Fe, ^{60}Co, ^{63}Ni, ^{125}Sb, ^{137}Cs. However, our high resolution γ-spectroscopy investigation revealed only the presence of ^{60}Co within the supplied material, in addition to ^{14}C and ^{3}H known to be present. The contact dose rate of the material was approximately 150 μSv h^{-1}, necessitating the use of lead shielding for extended manipulation procedures and storage of the material. To facilitate the active thermal treatment experiments, a purpose built vertical tube furnace was utilised, with a constant flow of air (120 or 240 ml min^{-1}) in order to carry through any volatilised ^{60}Co to be captured within a water scrubber or activated charcoal filter. However, no such release of ^{60}Co was detected from post-mortem γ-spectroscopy of scrubber liquor or filter material. Visual inspection and XRD analysis of thermal treatment experiments performed at 50wt% incorporation rate or below (120 ml min^{-1} air sparge), indicated that the process resulted in complete destruction of the resin feed, and afforded primarily a vitrified material with minor separation of a sulphate salt phase. SEM/EDX analysis of the vitrified material (Hitachi TM3030 instrument operating at 15kV), see Figure 2, revealed a homogeneous microstructure, with trace inclusions of an iron sulphide phase, which was expected given the high sulphur content and reducing nature of the melt. Further investigation demonstrated that an incorporation rate of 80wt% of dewatered resin could produce a slag like product with complete destruction of resin material, with an increased air sparge of 240 ml min^{-1}.

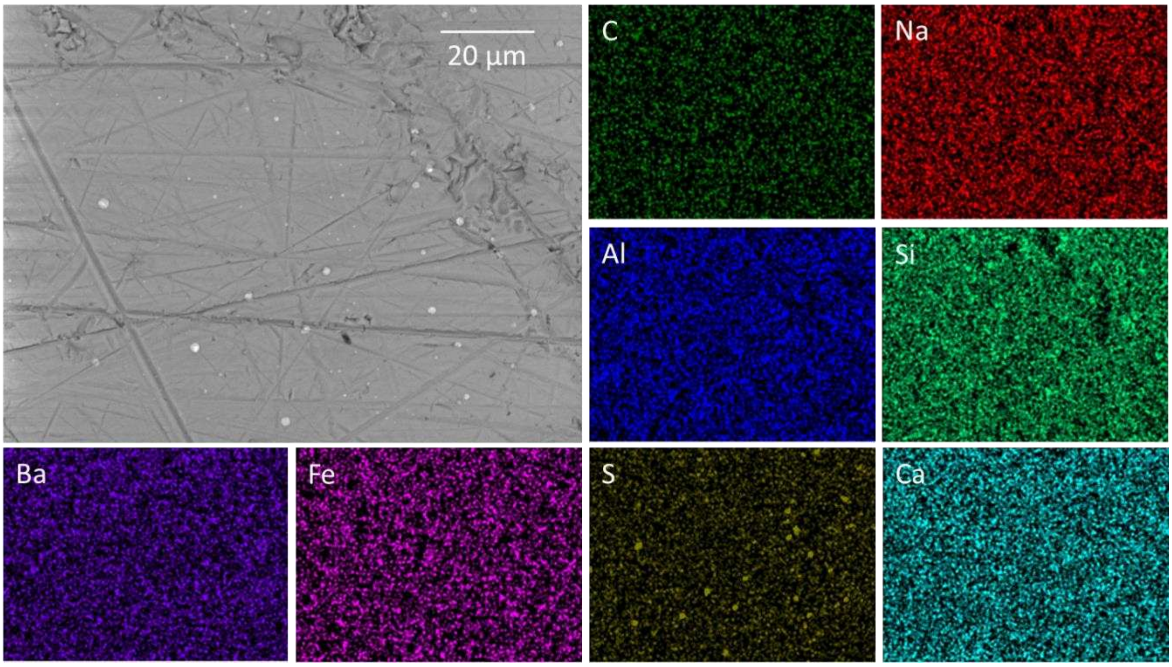

Figure 2. Backscattered electron image and elemental maps acquired from a representative area of the vitrified material obtained from thermal treatment of 50wt% real spent ion exchange resin (on de-watered basis) with G73 glass.

The process envelope evaluated at laboratory scale in this research is well within the application range of plasma vitrification technology, which is well suited to thermal treatment of organic radioactive wastes, such as resins and polymers [23, 24]. The conceptual products arising from thermal treatment of ion exchange resins are thought to meet the material related waste acceptance criteria of the Low Level Waste Repository. Therefore, the research summarised here has achieved the primary aim of demonstrating the efficacy of thermal treatment for resin destruction and production of a passively safe and homogeneous solid waste form, with minimal voidage, by destruction of the EDTA complexant and organic resin material. In particular, we emphasise the importance of validation of early stage surrogate research, using real radioactive waste resins, to develop early confidence in the compatibility of the approach.

5. Conclusions

The HADES Facility is established as a national centre of research excellence to support the UK nuclear decommissioning and disposal programme, in particular the development of thermal treatment technologies. Investment by The University of Sheffield, UK Government, and EPSRC has established state of the art capability and instrumentation for the handling of high inventories of radiological materials, including real radioactive wastes, to facilitate laboratory scale investigation of waste immobilisation technologies, characterisation of conceptual products, and evaluation of waste form disposability. The Facility provides unique capability within the UK academic landscape, including: handling of weighable quantities of ^{99}Tc and transuranics; quantitative electron probe microanalysis of radioactive materials; HIP of radioactive materials; laboratory-based X-ray absorption and emission spectroscopy. The HADES facility and equipment are open to external access via the arrangements of the UK National Nuclear User Facility and Royce Institute.

As shown in the case study of Section 4, the expertise and equipment established within the HADES Facility provide essential capability required to work with real radioactive wastes to develop and translate basic science and engineering into waste treatment processes, required to support strategic decision making. Such research capability is expected to play a pivotal role in accelerating the reduction in hazard, cost and timescale of UK nuclear decommissioning and enabling the case to be made for geological disposal of radioactive wastes, as set out in UK Government policy and the strategic plans of NDA and RWM.

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