

**Nuclear Decommissioning Authority
PhD Research Seminar for NDA-Sponsored PhD
Projects, 2018**

ABSTRACT BOOKLET

**17th January 2018
Manchester Conference Centre**

Nuclear Decommissioning Authority PhD Research Seminar for NDA-Sponsored PhD Projects

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Introduction

Rick Short (Research Manager, Nuclear Decommissioning Authority)

The NDA's mission to clean up the UK's civil nuclear legacy is going to take many decades and cost tens of billions of pounds. Over the years, we will need successive generations of highly skilled and knowledgeable people with the right experience to lead the technical aspects of the mission and help us and our supply chain to develop innovative solutions to clean-up challenges which can make the task safer and more economical. We also need to maintain a healthy academic body of expertise with a deep understanding of the decommissioning challenge. To help produce those future technical experts and industry leaders who are nurtured by the academic specialists, NDA spends over £500k per year directly sponsoring PhD students in the areas of characterisation, decommissioning, land quality, waste management and storage, spent fuel and nuclear materials.

This seminar is being held to encourage knowledge exchange between those with the industrial experience and those at the cutting edge of academic research in the decommissioning field. The students will present their work and have the opportunity to learn from the wealth of knowledge possessed by the industry experts present. Those experts from around the Estate and within the supply chain will get to learn about the tools and techniques that are being used in the academic community to address the clean-up challenges, and have the opportunity to help direct future research towards the challenges that they are uniquely placed to recognise.

With that in mind I'd like to thank you for attending the seminar and hope that you make the most of this opportunity to learn from each other throughout the day.

2018 NDA PhD SEMINAR PROGRAMME

Day 1: Tuesday 16th January 2018 - Early registration and poster drop off

16:00 – 18:00

EARLY REGISTRATION and POSTER DROP OFF

Day 2: Wednesday 17th January 2018 - Seminar Day

08:00 – 08:40

**REGISTRATION – Posters put up
COFFEE & NETWORKING**



08:40 – 08:50

WELCOME & INTRODUCTORY REMARKS

08:50 – 09:00

**NDA UNIVERSITY PROGRAMME
Rick Short - NDA**

09:00 – 09:15

NEW PhD STUDENT INTRODUCTIONS
(First year student will provide an introduction, max 4 minutes each)

**STUDENT
BURSARY YEAR - GROUP**

TITLE

Alana McNulty
2017 Bursary - Land Quality

Radionuclide alteration behaviour at the cement/subsurface interface: key controls on mobility

Alexandre Tribolet
2016 Bursary - Characterisation


Development of novel extractant test stick technology for the rapid screening of radionuclides

Malin Dixon Edwards
2016 Bursary - Spent Fuel and Nuclear Material



Brannerite glass-ceramic wasteforms for the immobilisation of nuclear fuel residues

Matthew Jackson
NGN - Spent Fuel and Nuclear Material

Long term storage of magnox fuel in dry conditions

09:15 – 09:55	PhD STUDENT POSTER INTRODUCTIONS (each 'poster presenter' will provide an oral introduction to their poster - max 4 minutes each To be followed by a poster session during lunch)
STUDENT BURSARY YEAR - GROUP	TITLE
Ikechukwu Ukaegbu 2015 iCASE - Characterisation	Radiological characterisation of hard to access areas: Integrated mixed field imaging and surface penetrating radar approach
Anita Crompton 2015 Bursary - Characterisation	Long-range scanning based detection of Alpha-Induced Air Fluorescence even under daylight conditions (AI-AF dc)
Richard Gray 2014 Bursary - Characterisation	Development & industrial testing of multi-radiation systems for characterisation of nuclear facilities
Danielle Merrikin 2016 Bursary - Characterisation	Element specific smart media for fast, low cost radionuclide analysis
Marco Boccaccio 2016 Bursary - Characterisation	Development of nonlinear acoustic techniques for the detection of cracks in storage containers
Diletta Invernizzi 2015 Bursary - Decommissioning	Benchmarking nuclear decommissioning
Cara Mulholland 2015 iCASE - Decommissioning	A qualitative approach to investigating the social implications of energy infrastructure: Building comparative case studies
Kate Lawrence 2014 iCASE - Decommissioning	Effective models for engaging and supporting SMEs in delivering technological innovation in nuclear decommissioning
Emma James 2016 Bursary - Land Quality	Assessment of the impact of leak zone processes in the natural attenuation of radionuclides
09:55 – 10:00	NDA Knowledge Hub Ronald Clark – National Nuclear Laboratory
10:00 – 10:45	COFFEE & NETWORKING 
10:45 – 11:20	PhD STUDENT POSTER INTRODUCTIONS CONTINUED (each 'poster presenter' will provide an oral introduction to their poster - max 4 minutes each To be followed by a poster session during lunch)

STUDENT BURSARY YEAR - GROUP	TITLE
Gianni Vettese 2015 Bursary - Land Quality	Optimising bio-remediation end point for the safe and long term stewardship of rad-waste
Chak-Hau Michael Tso 2014 Bursary - Land Quality	Enhancing the information content of geophysical data in nuclear site characterisation
Lewis Blackburn 2017 Bursary - Spent Fuel and Nuclear Material	Understanding the effectiveness of Plutonium surrogates for waste and stockpile immobilisation
Nathan Palmer 2014 iCASE - Spent Fuel and Nuclear Material	Computational modelling of PuO ₂ ageing and fuel residues
Jessica Higgins 2014 Bursary - Spent Fuel and Nuclear Material	Oxidation and radiation induced corrosion of uranium spent fuel
Sebastian Davies 2016 iCASE - Spent Fuel and Nuclear Material	The electrochemical treatment of nuclear wastes
Daniel Geddes 2016 Bursary - Waste Packaging & Storage	Design of a geopolymers cement for the UK nuclear industry
Patrick Pan 2017 Bursary - Waste Packaging & Storage	Remote magnetic monitoring of swelling in intermediate level waste canisters
11:20 – 12:00	KEYNOTE INDUSTRY TALK Adrian Bull – National Nuclear Laboratory Reflections on Nuclear Disaster – Lessons Learned (or not) as we Move on from Chernobyl and Fukushima
12:00 – 13:15	LUNCH AND POSTER SESSION (presenters to stand by posters from 12:30)
13:15 – 14:55	PhD STUDENT ORAL PRESENTATIONS (15 minutes presentation and 5 minutes Q&A each)

STUDENT BURSARY YEAR – GROUP	TITLE
Jamie Southworth 2013 iCASE - Spent Fuel and Nuclear Material	Investigation of anomalous H ₂ production from water adsorbed on metal oxides
Jack Clarke 2013 iCASE - Waste Packaging & Storage	Conditioning of legacy radioactive wastes requiring additional treatment
Jamie Purkis 2015 Bursary - Land Quality	Controlled Uranyl-Catalysed Hydrocarbon C-H bond cleavage
Adrian Cleary 2014 Bursary - Land Quality	Co-treatment of Sr and Tc in radioactively contaminated land with glycerol phosphate
David Hodkin 2013 Bursary - Land Quality	Enhanced crystallographic incorporation of Strontium(II) ions to calcite via preferential adsorption at obtuse sites during spiral growth
14:55 – 15:40	COFFEE & NETWORKING 
15:40 – 16:40	PhD STUDENT ORAL PRESENTATIONS CONTINUED (15 minutes presentation and 5 minutes Q&A each)
STUDENT BURSARY YEAR – GROUP	TITLE
Mel O'Leary DISTINCTIVE - Characterisation	Method for the determination of effective diffusivity and G-value of hydrogen in magnox sludge mimics
Kevin Tree NGN - Characterisation	Algorithm development for non destructive assay of radioactive waste
Robert Shearman 2014 Bursary - Characterisation	Using the National Nuclear Array as a fuel waste spectrometer
16:40 – 17:00	– closing remarks & presentation of prizes –
17:00	– Seminar Ends – Coffee is available 

NEW STUDENT INTRODUCTIONS

Radionuclide alteration behaviour at the cement/subsurface interface: Key controls on mobility

¹Alana E McNulty, ¹S.Shaw, ²L.Abrahamson-Mills, ¹G.Law and ¹K.Morris

¹University of Manchester

²National Nuclear Laboratory

Nuclear activities over the last 70 years, both military and civil, have left a global legacy of nuclear waste and radioactively contaminated land. Cementitious materials are used as a primary building material at nuclear facilities and play a large part in the immobilisation, storage and disposal of radioactive waste. Many nuclear facilities have significant quantity of radioactively contaminated land in-situ, and off-site disposal of this contaminated ground and building materials will be massively expensive. Here, we will explore of the speciation, fate and mobility of key radionuclides in model cementitious materials in sub-surface conditions. This will be achieved by a series of lab and field experiments to determine speciation and fate of radionuclides reacted with or encapsulated in cementitious materials relevant to radioactively contaminated land. Techniques we will use to characterise the radionuclide behaviour include radiochemistry, geochemistry, electron microscopy, luminescence spectroscopy, tomography and bulk and micro-focus absorption spectroscopy. Experimental findings on the cement/soil interface will ultimately be integrated into models to allow prediction of radionuclide mobility in the shallow sub-surface.

Development of novel extractant test stick technology for the rapid screening of radionuclides

¹Alexandre Tribolet, ¹P.E. Warwick, ¹I.W. Croudace and ²Steve Walters

¹University of Southampton

²National Nuclear Laboratory

Nuclear site decommissioning is underpinned by robust characterisation of the wastes arising. The UK and European nuclear industry are therefore investing in research into next generation characterisation technologies with exciting opportunities in this field. Screening of nuclear decommissioning wastes for non-gamma emitting radionuclides typically relies on time-consuming radiochemical analysis by expert laboratories remote from the decommissioning site. There are significant advantages to developing a rapid screening approach that can be deployed on site for the initial identification and quantification of radionuclides, permitting more time and cost-effective screening of wastes.

Working closely with industry and the Nuclear Decommissioning Authority, the project aims to develop novel analytical technologies for the robust, efficient and cost-effective characterisation on radioactive wastes. The research will focus on a purpose-designed extraction / detection system that can be deployed by non-specialists either on-site or as part of a mobile laboratory. The system will comprise three main components; (1) an optimised compact extraction system for recovery of radionuclides from the waste form; (2) an extraction test strip designed to recover the radionuclides from the waste digest; (3) a spatially-resolved, high efficiency digitized phosphor imaging system capable of quantification of alpha, low energy beta and high energy beta emitting radionuclides.

Brannerite glass ceramic wasteforms for the immobilisation of nuclear fuel residues

¹Malin Dixon Edwards, ¹Neil Hyatt and ²Ewan Maddrell

¹University of Sheffield

²National Nuclear Laboratory

Brannerite

The project seeks to investigate a possible glass-ceramic system with which to immobilise active wastes generated from damaged and degraded nuclear fuel materials on the Sellafield site. Brannerite is a naturally occurring uranium containing mineral ($U_2Ti_2O_6$), that has promising properties as a waste immobilisate. Over many millions of years natural brannerites become metamict, that is, fully amorphous as an effect of extended alpha-particle irradiation from their high uranium content. This metamictisation does not affect the uranium retention, which (if it translates to synthetic samples) is obviously desirable for a long-term wasteform. This, combined with the fact that natural brannerites can contain up to 4% water by weight, will hopefully allow for a glass-ceramic system that can be used in processing wet residues.

Hot isostatic pressing

Hot isostatic pressing is a high-specification densification process, where the material is sealed in a metal can before pressing under high temperature and pressure (up to 2000°C & 300 MPa). The synthesised brannerites will be HIPed along with a glass phase, in hopes of developing an excellent wasteform.

The inclusion of glass allows for partitioning of different active species, where long-lived U and Pu will be incorporated into the highly durable brannerite ceramic, and short-lived fission products (which make up around 3% of the targeted waste residue) incorporated into the less durable glass. If successful, this composite system should address the lack of waste treatment route for this material.



An example of the volume reduction achievable by hot isostatic pressing.



The University of Sheffield's research scale hot isostatic pressing facility.

References

- [1] G. R. Lumpkin *et al.*, *Chemical Geology*, vol. 291, Supplement C, pp. 55–68, Jan. 2012.
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Long term storage of magnox fuel in dry conditions

¹Matthew Jackson

¹University of Leeds

The aim of this project is to develop and test a process which will allow long term storage of Magnox Fuel in dry conditions, prior to disposal.

It is possible that a small quantity of Magnox fuel may not be reprocessed. Experience from the First Generation Fuel Storage ponds at Sellafield site shows us that wet storage of Magnox fuels is not possible for long periods. Therefore, an alternative solution is required for storage of Magnox fuel between reactor discharge and final disposal.

Wylfa has experience of storing fuel, freshly discharged from the reactor, in a purpose built dry store. Therefore technical feasibility for uncorroded fuel, with self heating, is proven; as long as the fuel remains dry. There are reports that if the fuel is accidentally wetted, then the cladding corrodes leading to the risk of activity release. If this is the case, then the sensitivity to even a short term exposure to water will greatly affect the possibility of long term storage.

The main question that this project will address is whether fuel that has already been stored and cooled in wet conditions can be dried and passivated sufficiently to allow long term storage without (further) degradation.

Some key aspects which must be considered for this project are:

- 1) Identify the likely cladding chemistry of wet stored Magnox fuel.
- 2) Recreate this cladding to establish surrogate materials for testing.
- 3) Determine the optimum physical conditions (pressure, temperature and time) for drying.
- 4) Determine the effect of gas composition on cladding chemistry.
- 5) Determine the effect of irradiation.
- 6) Investigate cladding stability during long term storage.

POSTER PRESENTATIONS

Radiological characterisation of hard to access areas: Integrated mixed field imaging and surface penetrating radar approach.

¹Ikechukwu K. Ukaegbu, ¹Kelum Gamage and ²Douglas Offin

¹Lancaster University

²National Nuclear Laboratory

Traditional methods of characterising wastes entrained in opaque materials e.g. soil and concrete, include: logging and micro drilling. These methods involve excavation which leads to the generation of secondary wastes and increases the risks of exposure of personnel and equipment to ionising radiation. Furthermore, non-destructive techniques such as radiation imaging techniques are only able to locate these wastes on the surface of the material in which they are entrained with no information on how deep the wastes are buried. However, ground penetrating radars (GPR) have proven to be a reliable non-destructive technique for locating and extracting material properties of objects located inside opaque materials. Therefore, this research aims to develop novel techniques for combining radiation imaging and GPR for improved characterisation of wastes entrained in different materials. To achieve this, a new depth profiling technique has been developed based on an approximate three-dimensional (3D) linear attenuation model. Results from simulation have shown that this technique has significant depth profiling ability compared to existing non-destructive techniques. However, the technique requires foreknowledge of the density of the material in which the radiation source is contained. This is usually not possible in typical field scenarios where the actual density of the material can vary significantly from standard calibrated values. Therefore, the new depth profiling technique is being further improved through the integration of real time density measurement from GPR signals. Another benefit of integrated radiation imaging and GPR radiological characterisation is the potential of non-destructive 3D reconstruction of the contaminated subsurface. Simulation results have demonstrated this ability in the case contaminated underground pipelines where the contaminated portion of the pipe were localised relative to other parts of the pipe by fusing radiation and GPR images. Such 3D images will be of immense benefit in guiding robotic systems during decommissioning. Finally, experiments are being undertaken to validate the simulation results for both gamma and neutron sources buried in different materials.

Long-range scanning based detection of Alpha-Induced Air Fluorescence even under daylight conditions

¹Anita Crompton and ¹Kelum Gamage

¹Lancaster University

Alpha particles ionise the surrounding atmosphere as they travel, causing fluorescence photons to be produced. These optical photons can be used to detect the source of the alpha emissions from a distance further than the mean free path of alpha particles, but so far this has mostly been achieved under special lighting conditions. This poster shows the primary results of experiments carried out to detect this fluorescence, with focus on photons in the UVC wavelength range (180 to 280 nm). A UVTron R9533 (Hamamatsu) sensor, designed to detect the UVC emissions from flames for fire alarm purposes or corona discharge, was tested in air in other gas atmospheres with a ²¹⁰Po alpha source to determine if this sensor had potential for stand-off alpha detection. The experiments were able to show that this detector is capable of detecting alpha-induced air-fluorescence in normal indoor lighting conditions. The gas atmospheres tested produced an increase in the detector count in general, with xenon having the greatest effect with a measured 52% increase in the detector response in comparison to the detector response in an air atmosphere. Nitrogen had little effect on fluorescence which may be due to oxygen quenching. The results show a positive outcome for the potential of this sensor for field alpha detection applications.

Development and industrial testing of multi-radiation systems for the characterisation of nuclear facilities

¹Richard Gray

¹University of Glasgow

The monitoring and characterisation of surface contaminated pipework poses difficulties for standard gamma spectrometers because the detector requires a large enough volume for full energy absorption, this may not be feasible in narrow pipelines. Novel, high density inorganic scintillators may provide a means to miniaturise our detectors while still maintaining a good energy resolution for an unambiguous radionuclide identification. Recent measurements using GAGG:Ce coupled to the j-series SiPM from SensL show promise and will be presented here. 9.9% energy resolution has been measured for the 662keV peak from Cs-137. This was achieved with a crystal just 3x3x30mm². The detector shows good linearity over its current dynamic range with predictable temperature characteristics.

Element specific smart media for fast, low cost radionuclide analysis

¹Danielle Merrikin, ¹Ian Fallis and ¹Simon Pope

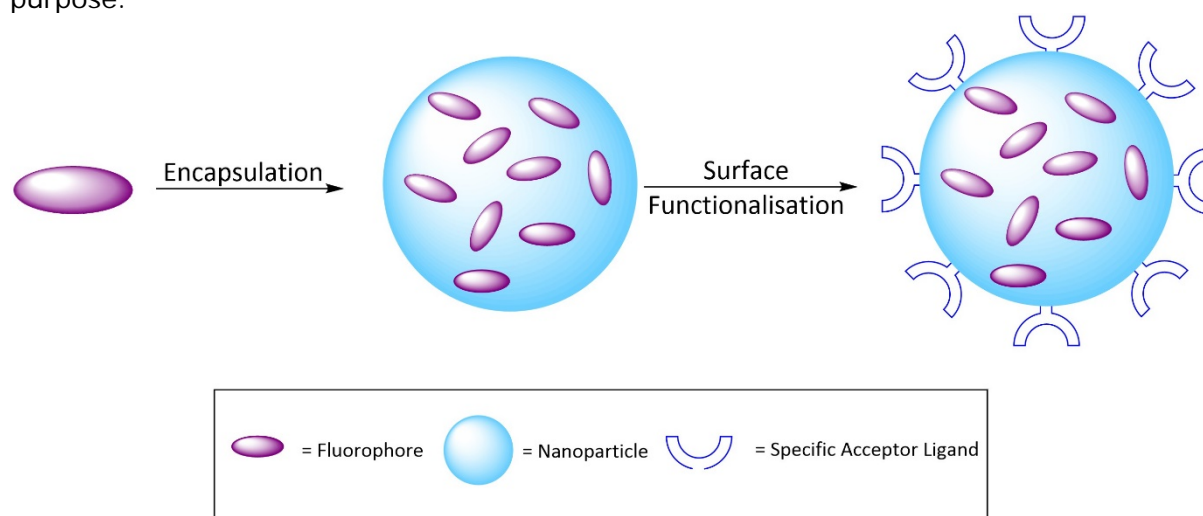
¹Cardiff University

Development of low-cost and time efficient analysis of low level waste (LLW) is an urgent need within the nuclear industry.

Liquid Scintillation Counting is currently employed in this analysis; however, this relies on harmful organic solvents, and as the samples are commonly aqueous the work-up of such samples is time-consuming. This not only has a large negative impact on the environment, but also increases disposal cost of samples.

For this reason, our approach utilises the Scintillation Proximity Assay, a method currently employed within the biological sector. Scintillant beads decorated with specific ligands on their surface can be introduced to an aqueous medium. The radioisotopes present within the sample will bind selectively to the bead surface, leading to emission of light following the interaction between the released beta particle of the radionuclide and the scintillant bead.

By exploiting this methodology, we aim to develop scintillant beads with surface functionality specific for a diverse range of radioactive nuclei commonly found in LLW, such as ⁹⁰Sr, ¹³⁷Cs, ⁵⁵Fe, and ⁶³Ni, allowing analysis of specific radionuclides in a complex mixture. We are investigating surface functionalised nanoparticle constructs for this purpose.



In the first 4 months of research a range of novel oxazole-based fluorophores, all with extended conjugation, have been developed using a method that is both low-cost and scalable. The derivatives made show a range of emission wavelengths with high quantum yields, suitable for scintillation. Strategies for doping of nanoparticles with these novel fluorophores is currently being investigated, utilising a green and low-cost methodology. Preliminary results exhibit similar steady state emission to the parent fluorophore, showing successful encapsulation.

Development of nonlinear acoustic techniques for the detection of cracks in storage containers

¹Marco Boccaccio

¹University of Bath

Conventional ultrasonic Non-Destructive Testing (NDT) monitors the scattering (reflection/refraction) of ultrasound waves with a defect, leading to amplitude and phase variations of the input signal due to the wave-defect interaction. The efficiency of classical ultrasound depends on the size of defects and a degradation of linear material properties (stiffness variation) caused by the damage. For incipient damage, the contribution of these factors is often very low and this determines the unacceptably low sensitivity of classical ultrasound technique for NDT for some type of defects/cracks.

The nonlinear acoustic approach proposed in this work makes use of the fact that cracks cause frequency changes of the input signal. These spectral changes are caused by anomalously nonlinear local dynamics of defects/cracks of various scale and nature. In cracked materials, the nonlinear response is provided by the Contact Acoustic Nonlinearity: strongly nonlinear local vibrations of cracks due to contact between crack surfaces, which generate multiple ultra-harmonics and multi-wave interactions. Basically, the intact parts of the material outside the defect vibrate linearly, i.e. with no frequency variation in the output spectrum, while a small cracked defect (invisible to a linear ultrasonic NDT) behaves as an active radiation source of new frequency components rather than a passive scatterer as in conventional ultrasound. By monitoring these nonlinear features a new class of NDT methods has been developed over the years able to detect early sign of deterioration in a number of materials. These methods will be investigated in our research and their field application will be studied and optimised for the detection and localisation of micro-cracks in storage cans and localised corrosion in spent AFR fuel cladding.

The nonlinear ultrasound methods are very sensitive to a numerous class of contact defects, scaled from dislocations (nano-scale) to fatigue (micro-) cracks etc. Since micro-crack defects are the forerunners of further major damage, the proposed nonlinear techniques are thus capable of early recognition of material degradation and "predicting" the oncoming fracture of waste packages during long-term storage. This makes nonlinear acousto/ultrasound methods proposed in this research a unique defect-selective instrument for localising and imaging of cracks in both storage cans and spent AGR fuel cladding.

Benchmarking nuclear decommissioning

¹Diletta Invernizzi

¹University of Leeds

Construction megaprojects are not new, and the body of knowledge in managing construction megaprojects has been developed by practitioners throughout the centuries and investigated by academics in the last few decades. Conversely, the body of knowledge on decommissioning is quite limited and project managers will need to focus more and more extensively on the challenges imposed by decommissioning projects.

Nuclear Decommissioning Projects and Programmes (NDPs) are complex, uncertain and expensive, often funded by governments and therefore politically sensitive, involving a large number of stakeholders, and sometimes socially problematic [1]. Decommissioning projects are also unique as no revenue-generating assets are created and there is no or little cash-in flow after the decommissioning project is completed: no “landmark output” is created and job positions are lost, without being replaced by new ones.

Benchmarking involves “*comparing actual or planned practices [...] to identify best practices, generate ideas for improvement*” [2] and offers significant potential to improve the performance of NDP selection, planning and delivery [3]. This research adapts a top-down benchmarking, developing a framework [4] to highlight the NDPs characteristics that mostly impact on the NDPs performance both through qualitative cross-comparison (e.g. [5]) and statistical analysis applied on European NDPs. This, however, needs to start with a clear understanding of “what a cost overrun really mean” [6], i.e. through a clear definition of how initial and end costs are assessed, the project stage these refer to, eventual scope changes, etc.

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A qualitative approach to investigating the social implications of energy infrastructure: Building comparative case studies

¹Cara Mulholland

¹University of Manchester

The complex social implications of an energy megaproject (a project with extensive costs, timescales, technical challenges and nationwide locations, [3] Flyvbjerg 2014) allow a context to push the theory of social value, which has been gaining increasing interest since the [6] Public Services (Social Value) Act 2012 promoting national discussions of social sustainability and influencing practices in infrastructure development. Major changes in UK energy infrastructure provide an opportunity to learn how to facilitate the transition to equitable, sustainable societies.

The lack of social science research in the energy field has been addressed by journals with calls for more research to understand the social implications [7] Sovacool et al. 2015. There is an emphasis on qualitative methods to investigate these issues in depth, bringing storytelling to the fore as a means of constructing meaning for the societies: "Narratives and stories allow people to connect social, economic, political, and technological elements together in ways that are meaningful to them" [5] Miller et al. 2015.

Interviews are an important source for rich primary data, which can be supported with observations. However, [2] Eisenhardt 2002 advocates for flexibility in research design, as each case needs to be explored in as much depth as feasibly possible, allowing for the addition of methods to capture information not found within the planned observations and semi-structured interviews.

To structure the research, [4] Knight et al. (2008) offer case studies as a useful research approach for the built environment, a sector largely driven by project based work. They provide guidance on undertaking case study research well within this context: provide a story for meaning in context; draw from multiple sources of evidence to triangulate; show in-depth understandings of the central issues, and relate to broader issues; have a clear focus; be reasonably bounded.

Case studies study phenomenon within its context using various data sources, with the boundaries between context and study being blurred, [8] Yin 2009. Case study theory building offers the opportunity to investigate evidence for new theories for a growing concept in a new context, and has been chosen because of the exploratory nature of this research [1] Eisenhardt & Graebner 2007, [2] Eisenhardt 2002.

References

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Effective models for engaging and supporting Small and Medium-sized Enterprises (SMEs) in delivering technological innovation in nuclear decommissioning

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²Nuclear Decommissioning Authority

Accessing an innovative idea from a small or medium-sized enterprise (SME) has received national and global recognition [1],[2],[3] and is of strategic importance to many public bodies. An innovative idea remains an 'opportunity' whilst navigating a path from concept to commercialisation. An innovative idea once applied – is an 'innovation'. However, from the perspective of a public body, the application of an innovative idea may simply be one of many innovation policy objectives. Objectives that are not exclusively associated with the deployment and commercialisation of an innovation. Objectives which can be impacted directly, or influenced indirectly, by the outcomes resulting from a public body's engagement mix. Setting a public body's engagement mix within the context of the public body's innovation policies provides a theoretical framing to analyse the factors that inform decisions on the engagement mix employed to access technological innovations from SMEs. Factors such as the role and capabilities of the public body. In respect to role, a public body as an "*innovation intermediary*" [4] between SMEs, the supply-chain and the wider knowledge network, is a contemporary research topic. Similarly, the capability to identify, assimilate and apply prior knowledge, the Absorptive Capacity [5] of a public body, is also an area of current academic interest. Therefore, through investigating how a public body engages with SMEs to access technological innovations, the research project addresses gaps within the academic literature pertaining to the public sector. Firstly, from the perspective of an unexplored form [6] of "*innovation intermediary*". Secondly, through viewing the ability of a public body to learn from, transfer and exploit previous experiences to engage with SMEs in terms of "*public sector Absorptive Capacity*".

A comparative case study method (utilising 'nested' project cases) is being followed. Data for the exploratory case-study (the NDA) has been obtained via semi-structured interviews and archival sources. Collected data is coded and analysed for key themes and patterns. Interim findings based on mapping the mix of SME engagement options utilised by the NDA in the context of the NDA's innovation policies, objectives and system activities [7] include: (a) the NDA facilitates innovation within sector, regional and national innovation systems through engagement options that broker knowledge transfer and networking opportunities between SMEs, the supply chain and wider knowledge networks; and (b) through 'intermediary' activities and influence, the NDA plays an active innovation systems role via engagement options which (1) stimulate a dynamic and innovative environment, (2) enable technology-transfer opportunities and (3) provide funding and access to R&D. This is an on-going research project. The current phase of data analysis involves the search for themes and patterns associated with the challenges, opportunities and measures associated with the mapped engagement mix. Following the completion of a comparative case study with an analogous UK public body, the value of the research project will be realised through contributing to the literature on public sector "*innovation intermediaries*" [6],[8],[9] and "*public sector Absorptive Capacity*" [10].

References

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Assessment of the impact of leak zone processes in the natural attenuation of radionuclides

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The Plutonium Uranium Redox EXtraction (PUREX) process and other similar spent fuel reprocessing procedures result in the production of nitric acid (HNO₃) based highly acidic-highly active liquors (HAL). Historical mismanagement of the long-term storage of these HAL has resulted in environmental contamination and the formation of subsurface acidic plumes at nuclear plants in the UK and USA; some of which have persisted for decades [1]. The NDA have identified that in some cases, the use of in-situ remediation techniques such as monitored natural attenuation (MNA) may be appropriate when managing contaminated land [2]. Thus, it is necessary to understand how the processes that naturally attenuate radionuclides are impacted when contaminants are introduced into the subsurface environment as part of a liquor with an extreme chemistry. The three objectives for this PhD have been briefly outlined in Figure 1.

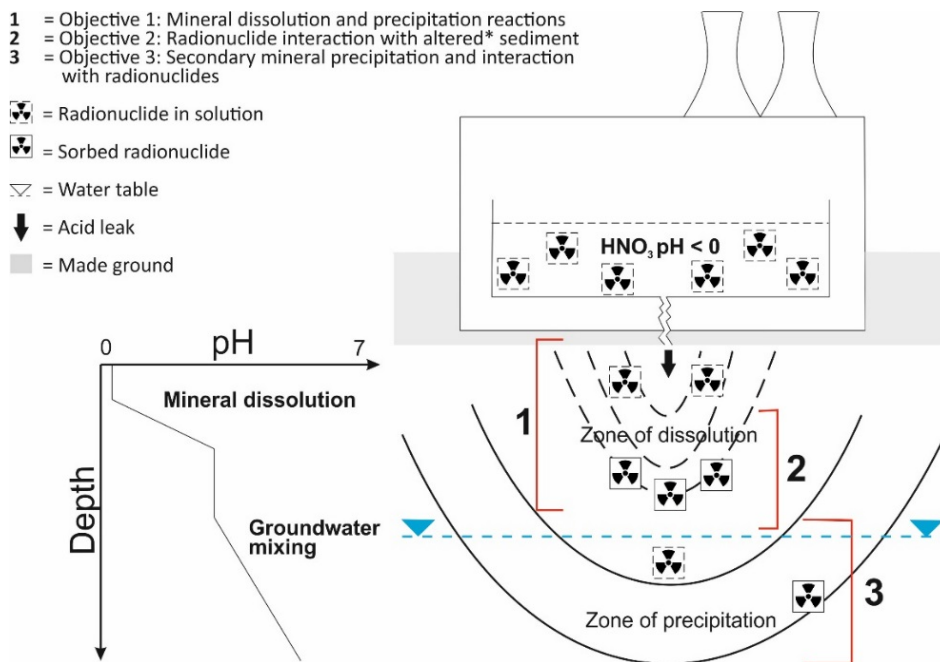


Figure 1. Conceptual model illustrating the key objectives of the PhD. *Altered sediment refers to sediment that has been chemically altered through exposure to HNO₃

Current work for this project involves the use of batch dissolution experiments coupled with ICP-OES, XRD, AFM and SEM to identify the mineral dissolution and precipitation reactions that occur when HNO₃ is introduced into the subsurface environment. Along side this, batch sorption experiments and surface complexation modeling (SCM) coupled with isothermal titration calorimetry (ITC) are being used to provide quantitative thermodynamic data for U(VI)-mineral binding reactions. These experiments are being conducted on unaltered and acid altered sediments/minerals, with the aim off assessing how U(VI) sorption to three Sellafield sediments is affected following prolonged exposure to HNO₃.

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Optimising bio-remediation end point for the safe and long term stewardship of rad-waste

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The mechanism of U(VI) bio reduction, a key bioremediation process

The addition of simple organic electron donors such as acetate has been shown to support the microbial reduction of soluble U(VI) to insoluble U(IV), immobilising the radionuclide in contaminated sediments. Although this “bioremediation” process has been reported widely, the mechanism(s) is not well understood. It is believed that the bioreduction of U(VI) by model Fe(III)-reducing bacteria such as *Geobacter spp.* and *Shewanella spp.* proceeds via a pentavalent uranyl(V) intermediate stage and that U(V) will be found in the aqueous supernatant during biotransformation, prior to disproportionation reactions that lead to the formation of U(IV) biominerals.

Earlier work has demonstrated enzymatic single-electron transfer to U(VI) species forming intermediate U(V) states in *Geobacter sulfurreducens*, [1] Jones et al. 2015; [3] Renshaw et al. 2005. We are widening the focus of this work to see if this mechanism is more widely distributed, using another model Fe(III)-reducing species *Shewanella oneidensis* MR-1. Here we provide an overview of data to date, including measurements made using fluorometry and XAS.

Long-term stabilities of co-treated sediments

Following new and promising results from the group, I am investigating the abilities of commercially available bio-remediation products for co-treating U and Sr; with special interest in sulphate rich systems as well as novel nano-scale Fe based treatments. Treatments include commercially available slow release electron donors, [2] Newsome et al. 2017 and a range of reactive nano and micron-scale materials, which will be compared to other well studied systems such as Ac/Lac electron donors and sulphate-reducing conditions.

This work will present clear and concise comparisons of several co-treatments for U and Sr contaminated sediments.

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Enhancing the information content of geophysical data in nuclear site characterisation

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Uncertainty in the subsurface characterisation of NDA sites poses significant risks in terms of operational cost and environmental protection. It is now recognised that improving our knowledge of the uncertainty of subsurface properties and processes is needed in order to enhance risk mitigation. Over the past few decades, geophysical methods have emerged as efficient and popular tools for hydrological studies. Compared to traditional point-based sampling methods, they have several unparalleled advantages, namely they: (i) offer high spatial and temporal coverage and resolution; (ii) are minimally invasive and easy to deploy; (iii) some methods are sensitive to changes in fluid properties; (iv) are cost-effective and not labour-intensive. A recent successful time-lapse electrical resistivity tomography (ERT) field trial conducted at Sellafield's Magnox Swarf Storage Silo (MSSS) highlights the potential of these methods, [1] Kuras et al. 2016. However, the relationship between geophysical signatures and the hydrological properties of interest can be complex and the former can be governed by multiple processes. Moreover, without hydrological information, the interpretation of geophysical images may not be hydrologically realistic. New methods are needed to incorporate hydrological information in the analysis of geophysical results, so that the resulting subsurface information can be judged in terms of its reliability, with realistic uncertainty bounds.

We illustrate the joint use of coupled hydrogeophysical modelling and data assimilation to effectively estimate flow and transport properties in leak plumes. Currently, geophysical inversion is the predominant way of interpreting ERT monitoring results. However, it is not an easy task to link images of geophysical properties to determine, for example, the mass discharge or flux of contaminant reaching a receptor. This has prevented more rapid uptake of geophysical methods in quantitative contaminated land risk assessment. Our approach avoids the use of time-consuming geophysical inversions. Instead, it proposes a range of hydrological models and then constrains them with time-lapse ERT data through data assimilation. As a result, we can obtain estimates of plume migration, with uncertainty bounds. The ensemble of hydrological model estimates also readily provides useful metrics for site management decisions, e.g. mass flux and mass discharge at any location or area within the model domain. We illustrate the proposed method by applying it to a simple vadose zone leak scenario (Figure 1) and a more complex Sellafield-like problem. The results demonstrate the reduction in uncertainty in plume mass discharge and estimation of leak rates.

We previously proposed an improved error model for geophysical measurements, [2] Tso et al., 2017 and showed that it can improve geophysical tomography results and uncertainty estimation. Ongoing work focusses on the role of uncertainty in petrophysical relationships required for estimating soil water content from geophysical tomography in the unsaturated zone. Overall, our work addresses NDA's needs by offering a suite of methods that can make geophysical methods more reliable, and hence lead to better decisions based on site characterisation results. Systematic application at NDA sites should contribute to a reduction in costs and risks in managing NDA's contaminated land portfolio.

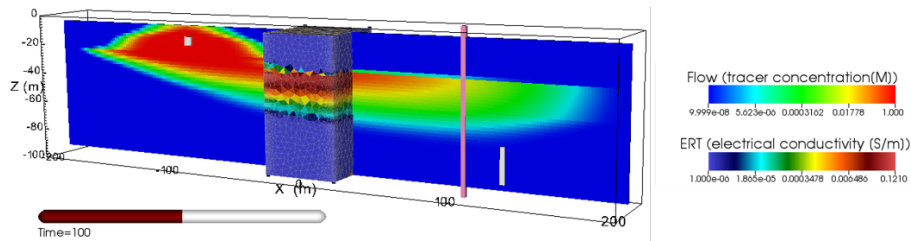


Figure 1: Combined hydrogeophysical model of a leak plume. In the 2D flow model, a leak plume (originates at the white dot) towards a production well in a regional aquifer (white line). The ERT imaging cell in the middle captures the changes in electrical conductivity induced by the plume. The pink line is a user-defined plane to calculate tracer mass discharge. The water table is at $z = -30\text{m}$.

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Understanding the effectiveness of Plutonium surrogates for waste and stockpile immobilisation

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There currently exists around 140 tonnes of separated PuO₂ contained at Sellafield as a result of reprocessing operations from civil nuclear power generation. Currently, the British government intends to reuse large amounts of the Pu as mixed oxide fuel (MOX) in the next generation of nuclear power plants, with prompt immobilisation and disposal of any portions of the stockpile deemed unsuitable for MOX fabrication. Candidate ceramic phases such as Zirconolite, CaZrTi₂O₇, have been previously shown to incorporate actinides at the atomic scale into their structure; as such show promise to immobilise the waste in a stable and chemically durable wasteform, fit for disposal in a geological disposal facility. The aim of this project is to understand the partitioning behaviour of Pu by the use of surrogates; small scale studies of this nature can demonstrate the transferability of knowledge by inactive studies. This will be achieved with the use of Hot Isostatic Pressing (HIP) technology, simultaneous application of high temperature and pressure under inert atmosphere, significantly reducing the volume of the waste package allowing greater throughput to a final repository.

Ionic radii and accessible oxidation states control the partitioning behaviour and solid solution mechanisms by which Pu can become incorporated into the ceramic structure. Uranium, Thorium and Cerium have all shown promise as surrogates for Plutonium; laboratory scale studies will use a variety of analytical techniques such as X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) to characterise the effectiveness of the surrogates. How well these formulations can mimic Plutonium behaviour will determine their future use in larger studies before final technical decisions are made regarding immobilising the stockpile. Systematic approaches to vary processing temperatures and controlled pO₂ will allow the effects of varying HIP conditions on the surrogate solubility and incorporation to also be investigated.

The conditions under which a particular surrogate will demonstrate behaviour alike to Pu will be investigated and this forms the main scope of the project. How HIP conditions can influence wasteform properties will allow the efficacy of this process to be characterised.

Computational modelling of PuO₂ ageing and fuel residues

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Background

The aim of this project is to use atomistic simulations to enhance understanding of PuO₂ in a storage environment. Hence, this work is very relevant to the UK's large plutonium (Pu) stockpile at Sellafield, Cumbria. Here, a brief summary of recent simulation results is given to help achieve the project aim. More specifically, the simulations are based on using robust interatomic potentials in static lattice and molecular dynamics simulations of PuO₂. These methods have enabled modelling of helium and defects in PuO₂. Indeed, helium behaviour in PuO₂ hasn't been investigated and is crucial to storage. This is because it is produced from the alpha decay of plutonium, also creating lattice damage, and possibly retained or released as gas in the storage canisters.

Static Lattice Simulations

Regarding static lattice simulations (using the GULP code), PuO₂ potentials from Arima et al. (2005) [1] and Read et al. (2014) [2] were used along with the Grimes et al. (1990) [3] potentials for helium modelling. Helium incorporation in lattice defects, clustering and migration has been investigated using the Mott-Littleton method [4] for defect calculations. Generally, analysis from the two potentials was concordant. Although helium interacts weakly in the PuO₂ lattice, it was found only single helium atoms are favourable in octahedral interstitial sites with a small exothermic incorporation energy average of -0.06 eV. A range of intrinsic defects were modelled as proposed helium trap sites, with helium incorporation energies calculated. Overall, the energy for a Schottky defect (a cluster of vacancies) was most exothermic at an average (total) of -0.64 eV, followed by oxygen vacancies at -0.41 eV. Figure 1 shows three helium atoms trapped in a Schottky defect in PuO₂. Migration of the helium atoms involving passage between adjacent oxygen ions gave rather high energy barriers of 4-5 eV, presenting a limiting factor for helium migration.

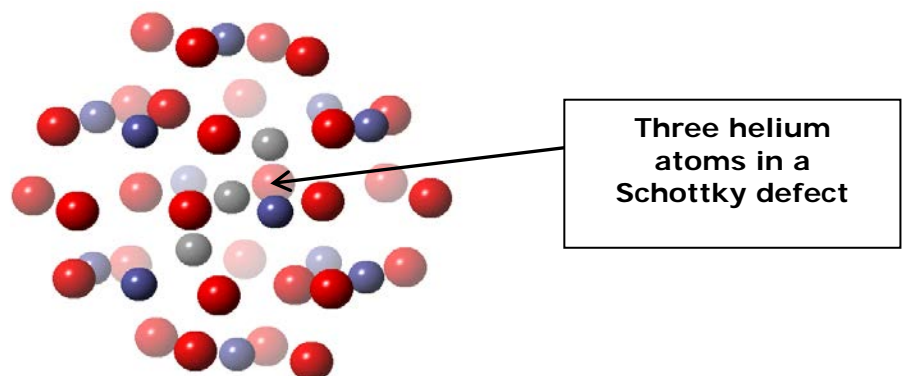


Figure1. A visualisation of three helium atoms trapped in a Schottky defect in PuO₂.

Molecular Dynamics Simulations

Molecular dynamics is a computational method in which the system (a collection of atoms) is evolved using Newton's laws of motion, to predict a range of dynamical properties. The simulations discussed used the DL_POLY programme. In addition to testing out the Read and Arima potentials for pure PuO₂, it has been used to model

helium behaviour and effects on thermal properties. The Arima and Grimes potentials were used. This was done using a 6 by 6 by 6 supercell of PuO₂, and insertion of helium atoms randomly in octahedral interstitial sites. The temperature was varied from 298-2400K and the helium atoms remained in these sites over the time of the simulations-typically 100ps, showing these sites are stable trapping sites for helium atoms. By varying the helium concentration, unexpectedly only up to 2% of the available space could be utilised. Hence, the effect on thermal properties, namely thermal expansion and heat capacity was very small. Although the concentrations are small, there is predicted to be an increase in the expansion coefficient with increasing helium concentration, increasing with temperature. The next phase of modelling will be to incorporate lattice defects to predict helium behaviour in defective PuO₂.

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Oxidation and radiation induced corrosion of uranium spent fuel

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During both the open and closed nuclear fuel cycles in the UK, spent uranium oxide fuel is initially placed into interim storage under alkali aqueous conditions. With spent fuel being an average of 97% uranium dioxide, the corrosion and dissolution of this ceramic matrix will be the primary release mechanism for other fission product species produced during in-reactor burn up. The interaction of radiation with liquid water has been widely studied and modelled with the stable products being hydrogen peroxide, gaseous hydrogen and oxygen. The yields of these products depend upon radiation quality, i.e. the radiation type and its energy. [1,2] Although the mechanism of for dissolution of uranium spent fuel under neutral aqueous conditions is known, and is defined as a corrosion process, the kinetics of these reactions remain largely unknown. [3]

Here we focus on the radiation induced chemical behaviour of water adsorbed onto the surface of spent uranium oxide fuels, within interim storage sites in the UK under gamma, high energy proton and 5.5 MeV helium ion irradiation. Initial studies we have examined include:

1. The oxidative dissolution mechanisms of the uranium oxide surface induced by reactive species produced in the radiolysis of water, and;
2. The quantification of these radiolytic products, in particular hydrogen peroxide, as a function of humidity, dose, dose rate, and stoichiometry.

The rate of formation of corrosion products such as studtite/metastudtite ($\text{UO}_3 \cdot y\text{H}_2\text{O}$) and speciation as a function of dose, pH and temperature within the $\text{UO}_2 \rightarrow \text{UO}_3 \cdot y\text{H}_2\text{O}$ system will be described along with the reaction kinetics obtained. We will show that spectroscopic analysis of the liquors can measure the chemical yields of hydrogen peroxide during irradiation of the $\text{UO}_2\text{-H}_2\text{O}$ system, while grazing incidence surface characterisation can probe the mineralogy of surface layers post-irradiation. [4]

A fuller understanding of this mechanism and the chemical interactions at the $\text{UO}_2\text{-H}_2\text{O}$ interface will form the foundations for a predictive model which will ultimately help to inform the safety case for long-term storage of spent nuclear fuels and this will be discussed further.

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The electrochemical treatment of nuclear wastes

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The nuclear industry has generated large amounts of waste materials contaminated with radioactive inorganic species. Some of these species, such as Plutonium (Pu) are particularly hazardous and often require very thorough decontamination techniques to separate the Pu compounds. The use of electrochemistry in processes can be a particularly elegant method of 'cleanly' achieving this objective without the need for chemical reagents and the associated, mass, volume and cost these bring. Reaction conditions and electrode potentials will be used to exploit the differences in thermodynamic stability of waste components to produce high degrees of selective separation. This will allow large reductions in the volume / mass of high hazard waste. The project will explore the fundamental electrochemistry of the proposed decontamination schemes, design and simulate the core electrochemical reactors involved and demonstrate their performance on a small scale. This will initially be with chemical analogues but there is an ambitious but achievable goal to use some active materials for direct demonstration of the process and reactor designs by the end of the project. This will be under very carefully controlled conditions at an industrial partner's site.

Design of a geopolymer cement for the UK nuclear industry

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Geopolymers are aluminosilicate based cementitious materials that can be used as an alternative binding system, to conventional blended Portland cements, for the immobilisation/encapsulation of wastes resulting from the nuclear fuel cycle. Geopolymers are formed from the chemical reaction between the aluminosilicate network of the precursor and an alkali activating solution, such as potassium silicate. These cements if formulated correctly, are highly fluid and as a result may give enhanced incorporation rates for waste types which may pose a rheological challenge for conventional Portland cements e.g. sludges and ion exchange resins. In addition, geopolymers also typically present a refined pore structure, along with tailorable chemical and physical properties suitable for the incorporation of problematic nuclear wastes such as oils and potentially reactive metals.

This study assesses the effects of mix design in the structural characteristics of metakaolin based geopolymer, particularly the effect of varying the amount of activating solution in the fresh state properties of geopolymers. To develop a system that can be of use to the UK nuclear industry, the fresh state properties of these cements need to be determined and understood, as they determine the procedure for the deployment of these cements within the waste disposal process. Rheological properties, setting time and kinetics of reaction of different geopolymer cements were determined, as well as their compressive strength development. Results are analysed based on key parameters and UK performance requirements for nuclear waste cementation, with the aim of identifying those geopolymers mixes that could be suitable for nuclear waste applications.

Remote magnetic monitoring of swelling in intermediate level waste canisters

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Nuclear energy is a necessity as a 'clean' energy source, which, as of 2017, produces a fifth (21%) of the UK's electricity [1]. However, nuclear power stations have been in operation since the 1940's and, until the enactment of the Energy Act in 2004 [2], there was little consideration of a sustainable strategy for nuclear waste disposal. As a result there has been a substantial accumulation of legacy waste that must be safely disposed of. The mission of the Nuclear Decommissioning Authority (NDA) is to develop strategies for safely and sustainably treating, monitoring, storing and ultimately disposing of both legacy and future nuclear waste, for the enormous timescales it remains hazardous [3].

In the UK, nuclear waste is typically generated from a "closed" operation fuel cycle, where the waste is divided to three levels; High Level Waste (HLW), Intermediate Level Waste (ILW) and Low Level Waste (LLW). Both HLW and ILW are wastes that contains a certain concentration of radioactive nuclides that remain from the fission reactions. To minimise contamination of the environment, these types of wastes are securely stored and isolated within canisters which are then placed at a licensed nuclear site for storage and monitoring. Typically, HLWs are stored using a vitrification process in borosilicate glass, whereas ILWs are contained in cement canisters [3]. This project focuses on the challenges associated with monitoring (in real time) swelling of the cement canisters for ILW.

Over timescales as short as a decade, a fraction of cement canisters will undergo swelling due to the production of hydrogen gas from the corrosion reaction between the ILW and the water in the cement [3]. It is important that this swelling is monitored to determine whether the canisters are stable enough for long-term disposal. Currently, visual inspections are used to monitor this phenomena, but there are approximately 44,000 existing ILW canisters [4]. Therefore, this technique is labour intensive and made difficult by the radiological hazardous environment. In this project we aim to develop an alternative, remote monitoring technique to detect the swelling by using ferromagnetic sensors.

This project will focus on developing a ribbon/wire of a strain-sensitive ferromagnetic material that can identify swelling of the ILW canisters by measuring their magnetic property, giant magnetoimpedance (GMI) (a phenomenon where electrical impedance - or resistance to AC current - is affected by changes in the magnetic states of ferromagnetic materials). We believe that once such a device is applied to the canisters, GMI measurements could be performed remotely, at a safe location, and allow users to distinguish the intensity of swelling in individual canisters. Here, we present progress towards the design of an experimental system for measuring GMI, and the development of a suitable sensor which could be utilised for the proposed application.

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ORAL PRESENTATIONS

Investigation of anomalous H₂ production from water absorbed on metal oxides

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In the UK, Legacy plutonium is stored in sealed canisters at the Sellafield site, Cumbria. Certain PuO₂ canisters have become pressurised over time, and a major component of the gas was determined to be H₂. The presence of a metal oxide can vastly alter the product distribution during the radiolysis of water. Most notably, for the radiolysis of water on e.g. ZrO₂, this includes an increase in the yield of H₂ by up to three orders of magnitude as compared to liquid water, whereas, the radiolysis of water adsorbed on PuO₂ yields lower quantities of H₂ compared to the radiolysis of pure water.

A novel experimental method for adsorbing water to the surface of metal oxides was developed. Replacing humidity control, a vacuum line technique was employed to introduce known quantities of water to the surface of dry oxide powders.

The γ -ray radiolysis of water adsorbed on ZnO or as an aqueous suspension was investigated as a surrogate for PuO₂ to establish the gas evolution. Surprisingly, both O₂ and H₂ were produced in similar quantities. This is unexpected as previously, either none or negligible volumes of O₂ are typically observed for water radiolysis on other oxides.

O₂ was observed during the radiolysis of both wet and dry ZnO, indicating the source of some of the O₂ to be the bulk oxide. The production of H₂ due to the radiolysis of water adsorbed on ZnO was observed to be an order of magnitude greater than for pure water for water adsorbed to the surface of the oxide. This could be attributed to an energy transfer process from the oxide to the adsorbed molecules. However, during the radiolysis of aqueous suspensions of ZnO the radiolytic yield of H₂ was observed to be an order of magnitude lower than pure water. Thus, ZnO has been seen to be a potentially suitable candidate as a surrogate for PuO₂ to investigate the anomalous effect PuO₂ has on the yield of H₂ during the radiolysis of water.

Conditioning of legacy radioactive wastes requiring additional treatment

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Intermediate level waste (ILW) (i.e. that which contains $>4\text{MBq kg}^{-1}$ α or $>12\text{MBq kg}^{-1}$ β/γ but generates $<2\text{kW m}^{-3}$ of heat) covers a wide range of waste materials found at nuclear sites. Thermal treatment offers several benefits for certain ILW streams including waste volume reduction and utilising the natural glass-formers present in the waste (such as SiO_2 and Al_2O_3). This poster shows results from laboratory-scale vitrification of two different ILW streams- problematic cemented Magnox fuel cladding and ion exchange resins. Various analytical techniques are used to characterise the resulting glassy wastefoms- including SEM/EDX (to study microstructure), XRD (for phase analysis), thermal analysis (to determine glass transition temperatures) and PCT-B (to study the durability of the glass).

Intermediate level waste (ILW) (i.e. that which contains $>4\text{MBq kg}^{-1}$ α or $>12\text{MBq kg}^{-1}$ β/γ , but generates $<2\text{kW m}^{-3}$ of heat) covers a wide range of waste materials found at nuclear sites. Thermal treatment offers several benefits for certain ILW streams including waste volume reduction. This effect can be amplified by utilising the natural glass-formers present in certain waste streams (such as SiO_2 and Al_2O_3) thus minimising the use of additional glass-formers. Additionally glassy materials are well-known to have a high chemical durability[1], particularly relative to other waste form materials.

This presentation will discuss results from laboratory-scale vitrification of different ILW streams- including problematic cemented Magnox fuel cladding, clinoptilolite ion exchange material and Magnox sludge. Borosilicate glass formulations are used with various analytical techniques utilised to characterise the glassy wastefoms including- SEM/EDX (to study microstructure), XRD (for phase analysis), thermal analysis (to determine glass transition and crystallisation temperatures) and PCT-B (to study the durability of the glass).

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Controlled Uranyl-Catalysed Hydrocarbon C-H bond cleavage

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The desire to better understand the chemistry involved in its safe storage has contributed to a renaissance in the field of actinide chemistry, with uranium and its compounds experiencing a myriad of exciting recent developments [1].

The irradiation of uranyl ($U^{VI}O_2^{2+}$) compounds with visible light to affect chemical transformations – uranyl photo-catalysis – is one such area. Shining light on solutions containing uranyl salts has been shown to result in the destruction of aqueous organic pollutants or the fluorination of inert C-H bonds [2]. Neither are easy chemical transformations to perform.

In aqueous solution, the photo-excited state of the uranyl ion can effect C-H bond cleavage by H-atom abstraction. We recently reported the first examples of hydrocarbon C-H bond cleavage *via* a thermal pathway [3].

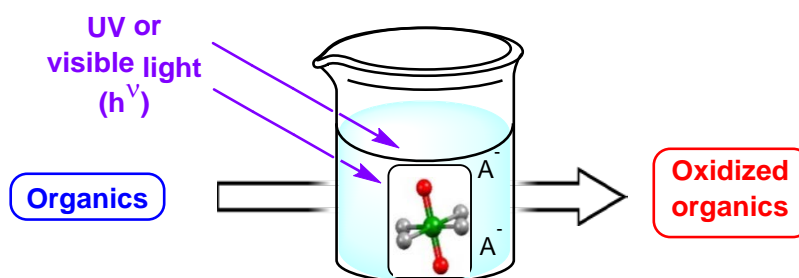


Figure 1: C-H bond oxidation using visible light uranyl catalyst

Targeting the room-temperature, environmentally-benign and chemo-selective uranyl photo-catalysed hydrocarbon transformation, we show that key transformations of common contaminants into industrially useful hydrocarbons is possible, with careful ligand design. We will present ongoing efforts to design uranyl photo-catalysts to target selective and challenging chemical transformations.

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Co-treatment of Sr and Tc in radioactively contaminated land with glycerol phosphate

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Nuclear fuel cycle operations over the last 60 years have resulted in a legacy of radioactively contaminated land at nuclear “mega-sites” such as Sellafield in the UK and Hanford and Oak Ridge in the USA. Uranium, strontium-90 and technetium-99 are all present at elevated concentrations in groundwaters at these sites¹. In-situ remediation technologies are currently receiving attention as a favourable means of treating the subsurface instead of “dig and dump” or traditional engineered barrier systems. It is important that these in situ approaches consider treatment of likely co-contaminants present at nuclear licensed sites and long term stability of the treatment products, and here we focus on Sr and Tc co-treatment.

Typically, the mobility of strontium in the environment is limited by sorption to clay minerals and Fe oxides; however sorbed strontium is exchangeable with other divalent cations, e.g. Mg^{2+} and Ca^{2+} and may be easily remobilised². By contrast, Sr^{2+} and Ca^{2+} bearing minerals such as apatite offer a long term, stable sink for Sr if mineralisation can be promoted³. Technetium exists under oxic conditions as the highly mobile pertechnetate anion ($Tc(VII)O_4^-$) although may be reduced to sparingly soluble Tc(IV) by reaction with Fe(II). Stimulating microbially induced Fe(III)-reducing conditions in the subsurface by the addition of an organic electron donor is therefore suggested as a means of immobilising technetium⁴.

The focus of this study is co-treatment of ⁹⁰Sr and ⁹⁹Tc using glycerol phosphate which has previously been shown to biomineralise soluble uranium to poorly soluble U-phosphate phases³. The project explores whether glycerol phosphate can stimulate formation of phosphate biominerals whilst also promoting development of Fe(III)-reducing conditions, causing biomineralisation of strontium and reduction of highly soluble Tc(VII) to poorly soluble Tc(IV). Initial experiments examined biomineralisation of Sr in a pure culture system of an environmentally relevant *Serratia* isolate⁵. The *Serratia* was grown anaerobically using glycerol phosphate in a synthetic freshwater minimal medium containing Sr^{2+} (as a proxy for ⁹⁰Sr) and Ca^{2+} . Following the biodegradation of glycerol phosphate, inorganic phosphate was released to solution and a decrease in Ca^{2+} and Sr^{2+} in solution was observed. Full characterisation included aqueous and solid phase analyses including XAS and these data will be discussed in the context of Sr treatment. Latterly, a series of sediment microcosms were then stimulated with glycerol phosphate under anaerobic conditions to examine Sr^{2+} and $Tc(VII)O_4^-$ behaviour in the microcosms. Here, results showed that sediments treated with glycerol phosphate removed significantly greater Sr from solution compared to control systems without glycerol phosphate. Tc removal was also substantial in the glycerol phosphate treated microcosms. Again, experimental data will be discussed in the context of co-treatment of Sr and Tc in contaminated land scenarios and geochemical, solid phase analyses and spectroscopy data will be used to support the conclusions.

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Enhanced crystallographic incorporation of Strontium(II) ions to calcite via preferential adsorption at obtuse sites during spiral growth

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⁹⁰Sr and ¹⁴C are two of the contaminants found at elevated levels in groundwater under the Separation Area of the Sellafield reprocessing site in Cumbria, UK. A conventional remediation technique for this suite of contaminants may involve pumping the groundwater and passing it through anion and cation exchange media. However, the effectiveness of this process is reduced by competing ions present in groundwaters (e.g. Ca²⁺, Na⁺)[1].

Precipitation of ⁹⁰Sr and ¹⁴C as insoluble carbonate [2] offers a potentially lower cost alternative, producing a solid residue that is readily grouted in cement wastefoms. It has been shown that this is a potentially viable technique with removal efficiencies of 99.7% of ¹⁴C and 98.6% of ⁹⁰Sr. Extrapolating this data to Sellafield site conditions suggests the effluent from this process would contain a residual 0.15 KBq L⁻¹ of ¹⁴C and 0.62 KBq L⁻¹ of ⁹⁰Sr [3]. However, Sr removal during this process displays two phases, with a slow initial phase followed by a faster secondary phase. In order to better understand the incorporation of ⁹⁰Sr into calcite a series of constant composition experiments were carried out under varying Sr:Ca ratios with calcite as the sole carbonate polymorph being precipitated.

These experiments displayed two calcite morphologies; a rhombic morphology (6.9-6 µm) which likely represents seed crystals, and a second elongate morphology (3.64-1.96 µm) which displayed an elongation along its C-axis with increasing Sr Wt. % of the precipitate. Low Sr/Ca precipitates displayed an XRD spectrum identical to that of rhombic calcite, however as the Sr Wt. % increased the calcite peaks shifted progressively to lower 2θ values, indicating increased lattice volume. Sr K-edge EXAFS analysis of the precipitates showed that the shift in morphology and lattice volume is accompanied by a change in the local coordination of Sr²⁺ in calcite. The Sr-O bond lengths were similar to the Ca-O bond lengths in calcite, but Sr-O coordination increased from 6 fold in crystals containing 0.21 Wt. % Sr, to 8 fold in crystals containing 9.47 Wt. % Sr, and the Sr-Ca coordination decreased from 6 and 6 (for the first and second Sr-Ca shells respectively) to 4 and 1.

It is suggested that Sr²⁺ is incorporated into calcite via incorporation into the lattice (rather than sorption) and that it undergoes preferential incorporation at obtuse (+) growth sites on the calcite surface due to its large ionic radius (1.13 Å). This would lead to the observed elongation parallel to the C-axis. It is therefore apparent that Sr²⁺ is compatible in the calcite lattice across a range of Sr:Ca values (although this introduces a degree of disorder therein).

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Method for the determination of effective diffusivity and G-value of hydrogen in magnox sludge mimics

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This project investigates radiation-induced processes involving Magnox nuclear waste sludges, which are poorly characterised, attempting to expose the underlying mechanisms at work in the sludges to inform the current and future handling of these sludges. The processes being investigated involve the breakdown through irradiation constituents of the sludge to produce hydrogen. In the sludges radiation is due to the decay of radioactive materials present. Mg(OH)₂-water mixtures were irradiated to mimic the properties of sludges and to provide an insight into the mechanisms at work. The two properties measured were the effective hydrogen diffusivity in the sludge mimic and the radiolytic yield of hydrogen in the sludge mimic (G-value). The hydrogen concentration is measured at a known distance from an irradiated region, with a Unisense hydrogen microsensor. The distance from the probe to the irradiated region shorter than the width of the irradiated region. This means that the diffusion can be modelled as a 1-dimensional problem, which has an analytic solution[1]. This means that the concentration change over time can be fitted to this model. From the best fit the diffusivity and G-value can be calculated. From this analysis, the effective diffusivity of hydrogen through the sludge mimic was determined for a simple sludge mimic comprising 10 nm Mg(OH)₂ nano-particles mixed with the water (0.2 w/w Mg(OH)₂) with 20 keV x-rays, see figure below. The hydrogen G-value can also be calculated from this model. This method was also applied to at other energies and different mimics.

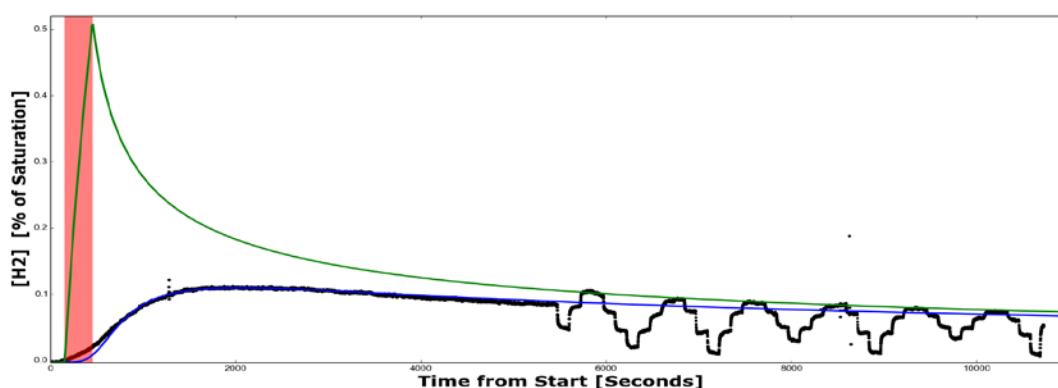


Figure: Results from 10 nm Mg(OH)₂ nanopowder mixed with water (0.2 w/w Mg(OH)₂). The with the probe stationary 2 mm above the irradiation region, and then with the probe scanning up and down through the sample. The blue line is the 1D model fit to the data giving: hydrogen diffusivity in sludge relative to in water of 0.444 ± 0.002 and G-value of 0.043 ± 0.007 $\mu\text{mol}/\text{J}$. The green line represents the predicted concentration of hydrogen in the irradiated region, based on the fit. The red region on the plot shows when the irradiation took place.

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Algorithm development for non destructive assay of radioactive waste

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Broad Energy Germanium detectors are routinely used for gamma ray spectroscopic analysis of nuclear waste streams to ascertain the type and relative activity of radionuclides. Challenges arise when low activity radionuclides of interest are obscured by the presence of higher activity radionuclides. A key example is when Compton scattered gamma rays from ¹³⁷Cs in the spectrum conceal the presence of low energy gamma rays from ²⁴¹Am. The Compton scattering events manifest in a continuum within the spectrum, elevating noise. Current methods to overcome this problem involve the use of Compton suppression systems that use a central broad energy germanium detector surrounded by a guard ring of bismuth germanium oxide scintillator detectors. Any gamma-rays which Compton scatter from the germanium detector and then interact in the scintillator detectors will be vetoed from the germanium detector energy spectrum using timing coincidence methods. In this project, algorithms have been developed to perform digital Compton suppression, alleviating the requirement for scintillator detectors. This has been achieved by distinguishing low energy gamma rays that are absorbed near the outer surface of the germanium detector, from Compton scattering events that are as a result of higher energy gamma rays interacting throughout the bulk of the detector. The digital Compton suppression algorithm significantly reduces counting times with improved low energy isotope identification when in the presence of large backgrounds, without the requirement for Compton suppression shielding. It also allows for a "one detector" system to be utilised. The technique exploits the knowledge of how the detector signal varies as a function of gamma-ray interaction position, characterised by position-dependent charge collection times. Databases of experimental and modelled detector signals have been produced for known (x,y,z) gamma-ray interaction positions in the germanium detector. The modelled signals were produced using the AGATA Detector Library and the experimental signals were collected through precision scanning of the detector. A 1GBq ¹³⁷Cs source calibrated into a 1mm beam was mounted on a software controlled (x,y) scanning table below the germanium detector. As the collimator was moved, the detector was thus irradiated at known (x,y) positions. To determine the depth of interaction within the germanium detector, scintillator detectors were positioned behind collimators at various known z depths alongside the germanium detector. The position dependence of charge collection times could then be investigated. In this paper, the background to the project together with its aims will be presented. Presented results will include the dependence of charge collection times on both radius and depth (z) within the Broad Energy Germanium detector. Additionally, the results of the optimised Compton suppression algorithm will be presented. The results show that the Compton suppression algorithm is successful and has enabled a reduction in the Minimum Detectable Activity of the detector. It is envisaged that future research to implement the algorithm in real-time will be conducted. Real time application will allow for a portable single detector system to be used by waste producers. This will allow waste producers to perform efficient characterisation of low level nuclear waste with confidence. The

reduction in the minimum detectable activity will enable waste producers to re-characterise some of their low level waste as very low level waste. Very low level waste can be diverted to licensed land fill sites rather than being sent to the low level waste repository.

Using the National Nuclear Array as a fuel waste spectrometer

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The National Nuclear Array (NANA) at the National Physical Laboratory (NPL) has been used to determine the specific activity, the activity per unit mass of material, of isotopes where the decay is succeeded by a gamma cascade [1]. This, merged with previous work [2] utilising the modularity and timing precision of lanthanum tri-bromide detectors, to identify species according to these signature, coincident and prompt gamma-rays can be theoretically used to identify and quantify the amount of certain radioactive products within a waste package. The concentration of nuclides in waste is an essential piece of knowledge for the development of the procedures to decommission radiologically contaminated materials. This talk will show the work from this thesis regarding NANA: the clean selection of specific radionuclides within a prepared sample of ¹³⁴Cs and ¹³⁷Cs which are important fission product wastes in spent nuclear fuel. The talk will also outline the methodology and the results and steps taken to improve the result of using NANA to calculate the specific activity of ⁶⁰Co and an assessment made of the spectrometer in relation to the more formalised and oft-used method of digital coincident $4\pi\beta$ counting [3]. Finally, a GEANT4 [4, 5] simulation-based investigation into using NANA to assay high-active separated plutonium powder from reprocessing will be presented. Comparisons with the current technology, a single large Thallium-doped Sodium Iodide will be made.

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