

# DISTINCTIVE 2017

3<sup>rd</sup> Annual Meeting, York, 5th - 6th April

DISTINCTIVE  
Decommissioning,  
Immobilisation and  
Storage soluTions for  
NuClear wasTe InVentories  
A university consortium funded  
by the Research Councils  
UK Energy programme



**A university consortium funded by the:**



**Thank you to our key project partners for their support and significant contributions to the programme:**



**A special thank you to our event sponsors:**



## Welcome

On behalf of the DISTINCTIVE Leadership Team, I welcome you to the National Railway Museum, York, for the 3<sup>rd</sup> Annual Meeting of the University Consortium.

The 1<sup>st</sup> conference was held at the Millennium Gallery in Sheffield in April 2015, with the second at the At-Bristol Science Centre, Bristol in April of last year. Both events attracted about 120 delegates with equal representation from academia, industry and associated stakeholders. These meetings allowed all organisations associated with the project to come together, and were useful in enabling valuable interactions for mutual benefit and significantly raised awareness of the programme within the waste management and decommissioning community.

These meetings form a significant component of our impact activities for beneficiaries in site licence companies and the associated industrial supply chain, together with the technical theme meetings held annually in October/November. We are also pursuing similar dissemination activities through previous and planned industry roadshows, and at key national and international conferences, as well as through a number of challenge-led meetings on specific industry needs-led research challenges planned for the coming year.

We have continued to engage society and stakeholder groups through public engagement events held, in one instance, as part of the Bristol European Green Capital Programme, and recently held a three-day public engagement and media summer school for our PhD students and PDRAs to train them in public engagement, media and science writing. We are also planning a project documentary for use online and at science festivals, and a World Café targeted at National Science Week.

We have been successful in influencing government, regulators and implementation authorities, through representation at the All Party Parliamentary Group on Nuclear Energy to discuss research on immobilisation of plutonium residues and stockpile material, through membership of the Radioactive Waste Management working group on fuel disposal and behaviour, and in one case as a Special Advisor to the House of Lords to assist with running the Science and Technology Committee investigation into the future of UK nuclear R&D. Our academics have also presented to different audiences in the US and Japan as part of HMG Foreign & Commonwealth Office delegations to potential UK partners overseas.

Time flies and we are now two-thirds of the way through the programme, with many of our researchers beginning to think about their future careers. DIAMOND, DISTINCTIVE's predecessor, was very effective in retaining researchers within the nuclear sector, and it was through events like this that initial prospects were discussed, and we encourage both our researchers and industry colleagues to take advantage of the opportunities presented by this meeting. We are having four employability-related presentations as part of the meeting

from nuclear researchers who went on to make careers in both academia and industry, and a number of prospective employers will be exhibiting during the meeting.

I look forward to introducing you to our Invited Speakers, Prof. Ian Pegg (Catholic University of America and chair of our International Advisory Group), Dr. Albert Kruger (US Department of Energy) and Andrew Cooney and Dr. Stephen Hepworth (Sellafield Ltd.), and thank them for contributing to this event.

I would also like to take this opportunity to acknowledge funding from the Research Councils UK Energy Programme (EP/L014041/1), and to thank our key project partners, NNL, NDA and Sellafield Ltd., for their continued support and contributions.

Lastly, I would like to say a special thank you to our event sponsors, Longenecker & Associates and the NDA, for their sponsorship of this event.

A handwritten signature in blue ink, appearing to be 'M. Fairweather', with a stylized flourish at the end.

Prof. Michael Fairweather (Principal Investigator – [M. Fairweather@leeds.ac.uk](mailto:M. Fairweather@leeds.ac.uk))

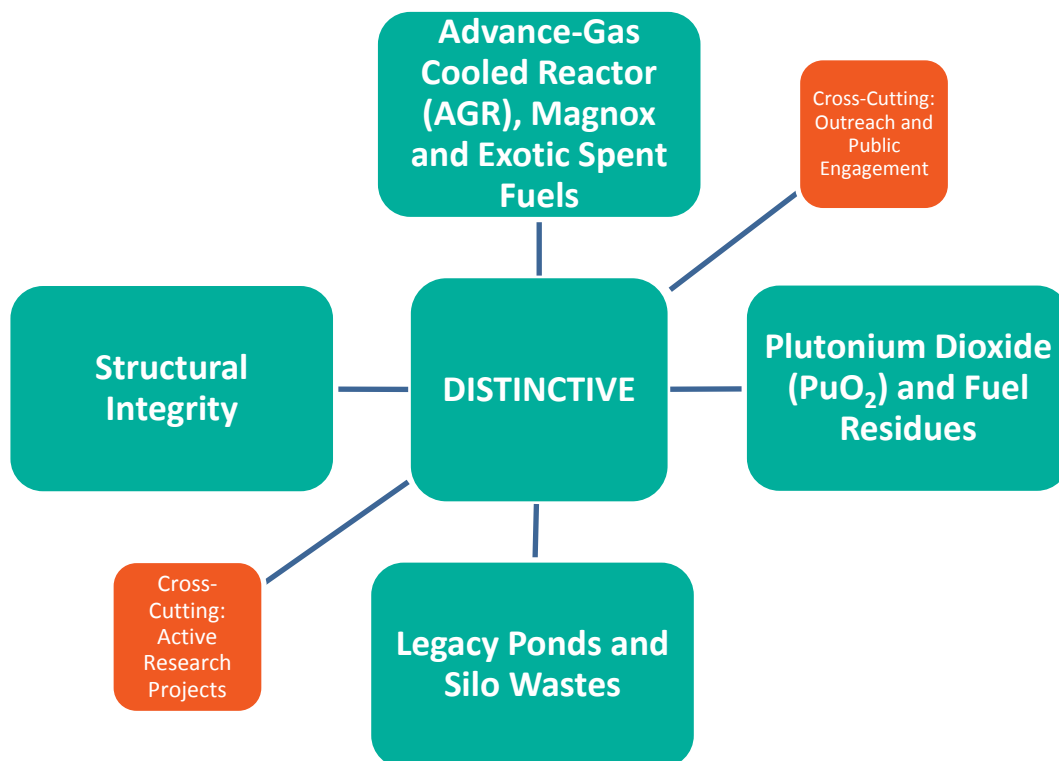
## Programme Overview

The structure of this world-class research programme has been aligned with the strategic needs of the UK industry in the area of nuclear waste management and decommissioning.

The aims of the consortium are:

- To carry out internationally leading science and engineering research in the area of decommissioning and nuclear waste management.
- To support research that provides routes to innovative technology developments that can be applied to decommissioning and nuclear waste management.
- To foster and develop new multi-disciplinary research partnerships between academic and industry researchers.
- To train the next generation of UK researchers, equipping them with skills and experiences relevant to nuclear waste management and decommissioning issues.
- To provide a focal point for government, industry and academics through which current and future R&D issues associated with nuclear waste and decommissioning can be discussed.
- To provide a route for public understanding of the underlying research and development needs opportunities and solutions to nuclear waste and decommissioning.

All **53** research projects fall into one of four technical themes which were co-identified with our industrial partners; NNL, NDA and Sellafield Ltd.



## Theme 1 - AGR, Magnox and Exotic Spent Fuels

**Aim:** To provide technical underpinning to the options for the management of the UK's AGR, Magnox and Exotic Spent Fuels

**Objectives:**

- To understand the evolution of Magnox and exotic SNF during recovery from aqueous storage, drying and repackaging.
- To develop spectroscopic methods for improved determination of SNF dissolution and corrosion rates in water.
- To determine the optimum drying conditions for AGR fuels and the subsequent surface reactivity and alteration of unclad  $\text{UO}_2$  in dry storage.
- To determine the consequences of radiation damage in SNF, cladding and other wasteforms for safe long term storage.
- To determine suitable waste management options for spent carbide fuels.

## Theme 2 – $\text{PuO}_2$ and Fuel Residues

**Aim:** To provide technical underpinning to the options for the UK's civil Plutonium inventory

**Objectives:**

- To understand how the structure and properties of  $\text{PuO}_2$  change with time in the presence of  $\text{H}_2\text{O}$ .
- To understand the roles these processes play in gaseous product evolution from  $\text{PuO}_2$  in storage.
- To understand radiation induced amorphisation and dissolution kinetics of Pu wasteforms.
- To develop novel, fast neutron based radiometric methods for the quantification, isotopic composition assessment and remote imaging of Pu bearing materials.

## Theme 3 – Legacy Ponds and Silo Wastes

**Aim:** To develop innovative technical approaches to clean up UK legacy wastes.

**Objectives:**

- To understand durability of heterogeneous ILW glass/ceramic wasteforms from LP&S wastestreams.
- To develop improved ways to remove radionuclides (RNs) from solution, both novel inorganic ion exchange solids and tailored binding superparamagnetic nanoparticles, to treat complex and variable effluents.
- To develop new micro- and ultra-filtration methods for use with sludges.
- To provide three-dimensional modelling and simulation for sludge disturbance, mobilisation and transport, with supportive experimental studies, and manipulation planning for removing corroding nuclear materials.

- To develop a better understanding of gas hold-up in sludges.
- To develop improved techniques for remote monitoring of sludges and heterogeneous wastes.

## Theme 4 – Structural Integrity

**Aim:** To develop reliable systems for infrastructure characterisation, restoration and preservation, that minimise current, and future, radiation exposure to the workforce whilst providing economically viable technological solutions.

### Objectives:

- To develop in-situ ground barriers that could act as a ‘second skin’ surrounding on-site structures, such as silos and ponds, for prevention of subsurface radionuclide migration.
- To develop smart solutions for remote crack detection, infrastructure health prediction and building preservation that can be retrofitted to existing sites.
- To develop autonomous systems with increased functionality and to coordinate them through a CAD-based real-time management system, to facilitate planning and execution of decommissioning works.

## Cross-Cutting Themes

The Leadership Team will continue to identify common themes across the research projects to maximise collaboration, training and knowledge/technology transfer across the consortium.

Two cross-cutting themes currently exist; Active Research Projects, and Outreach and Public Engagement.

### Active Research Projects

A key component of the DISTINCTIVE programme is the use of world-class active research facilities, both within the UK and internationally. Prof. Simon Pimblott is responsible for ensuring that the consortium receives excellent advice and support in this area, especially relating to the technical needs and duration of the work.

### Outreach and Public Engagement

The consortium has identified three distinct groups of non-academic beneficiaries from consortium activities; site license companies and the associated industrial supply chain; Government, regulators and implementation authorities; and, society and stakeholders groups. Over the duration of the programme, DISTINCTIVE will deliver a variety of activities, led by Prof. Neil Hyatt, which will have impact in four key domains:

- Knowledge
- People
- Economy
- Society

## Introducing the Invited Speakers

### **Prof. Ian I. Pegg (Catholic University of America, USA)**



Dr. Ian L. Pegg is Professor of Physics and Director of the Vitreous State Laboratory (VSL) at The Catholic University of America. His research has spanned various areas of materials science including the optimization of processes and glass compositions for use in nuclear waste disposal, geopolymers, nano-materials, and thermoelectrics. Dr. Pegg has led numerous vitrification R&D programs involving the development and characterization of glass formulations and the demonstration and scale-up of Joule-heated melting processes. Dr. Pegg has served as a technical team member on several successful multi-billion-dollar treatment facility proposals, including the WTP privatization at Hanford and the AMWTP privatization at Idaho. Dr. Pegg directs the Hanford WTP vitrification research and technology support effort at VSL, which supports the design, construction, and operation of what will be the world's largest nuclear waste vitrification facility. Dr. Pegg also directs similar R&D efforts for the Defense Waste Processing Facility at the Savannah River Site and for the Rokkasho vitrification facility at Rokkasho, Japan. Dr. Pegg has been a frequent participant on expert review teams for the US Department of Energy. He is the co-founder of three advanced materials companies. He previously held positions at the National Institute for Standards and Technology and in the Department of Chemistry and Biochemistry at UCLA. He holds a PhD in physical chemistry from The University of Sheffield as well as MBA and BSc degrees.

### **Andrew Cooney (Sellafield Ltd)**



Nuclear Professional with over 25 years of experience in the industry, especially decommissioning and high hazard facilities. I have a Degree in Physics and strong technical background which includes experience in technical, safety and environmental management, safety case and strategy roles. Currently working in the Sellafield Strategy and Technical group on external innovation and with NNL on academic programmes.



### **Dr. Albert A. Kruger (DOE Office of River Protection)**



Albert joined the staff of the Office of River Protection in 2007 after working in the laboratory of the world's oldest and largest glass company, Saint-Gobain Recherche in Aubervilliers France, 3M Central Research Physic & Materials Group, and Bell Telephone Laboratories. In May 2008 he was selected as the Glass Scientist and in December of 2010 he was named as Supervisor of the Vitrification Group. Albert has responsibility for glass formulation, glass durability testing, vitrification process capacity efficiency and involvement with the melters. The fruits of his thoughts, labors and collaborative efforts funded by DOE have produced the very real possibility that the Hanford Waste Treatment and Immobilization Project will be significantly more efficient and flexible in meeting mission success criteria. These improvements include higher waste loadings for low and high activity waste forms, significantly higher throughputs in the existing facility designs, and greater flexibility with flowsheet and feed vector options.

When it existed, he was responsible for the interface with the disposal authority as it related to waste form conformance to disposal requirements for high-level waste. Of note, in Albert's first three years with ORP, he and his Bechtel colleague were the first to prepare and submit an IHLW Waste Form Compliance Plan and subsequently an IHLW Waste Form Qualification Report and obtain EM-HQ and DOE-RW approval in 3 decades of attempts.

Albert holds in excess of 30 patents world-wide in glass science, has authored and presented over 100 scholarly papers including a prestigious Solvay Seminar, and lectured at the NATO Summer School on Oxide Surfaces. Albert is an Eagle Scout, recipient of the Silver Beaver of the Boy Scouts of America, Fellow of the American Institute of Chemists and a Full Member of Sigma Xi.

### **Dr Stephen Hepworth (Sellafield Ltd)**



Dr Hepworth is a physicist with over 20 years of experience in radiation measurement and nuclear decommissioning technology development. Steve is the Research, Development and Innovation Programme Lead of Sellafield Ltd.'s Strategy and Technical Directorate. His experience includes serving as the Sellafield Head of Technical Standards and Assurance, and as the Lead of a multi-disciplinary study into a groundwater management scheme of the UK's most hazardous nuclear facility. Throughout his career he has been influential in R&D across the nuclear decommissioning spectrum including modelling and simulation, synthetic environments, radiometric imaging, robotics, aerial vehicles, decontamination, cutting technology and semi-remote tooling. He is Fellow of the Institute of Physics and is the physics graduate scheme leader at Sellafield.

## PhD Student Awards

A £250 prize will be awarded for the best oral and the best poster presentation given by a DISTINCTIVE PhD student.


Our industry delegates (only) are invited to register their vote at the Registration Desk by 15:30 on Thursday 6th April. A voting sheet can be collected from the desk (or Lois Tovey) at any time.

The awards for each category will be presented by Dr. Rick Short on behalf of the NDA, who kindly sponsored these awards, at the end of the event.



# Agenda

Wednesday 5 <sup>th</sup> April		
Time	Title	Presenter
10:00	Registration / Poster Board set up / Refreshments	
11:00	Introduction and Housekeeping	Prof. Michael Fairweather  University of Leeds
<b>Session 1</b> <i>Chairs: Prof. Neil Hyatt, University of Sheffield &amp; Prof. Simon Pimblott, The University of Manchester</i>		
11:10	Key findings of DISTINCTIVE to date	Prof. Ian Pegg Catholic University of America Andrew Cooney Sellafield Ltd
11:50	Insights into the corrosion behaviour of uranium SNF using thin films	Dr. James Darnbrough,  University of Bristol
12:10	Water Layers on Actinide Oxide Surfaces	Dr. Bengt Tegner,  University of Manchester
12:30	Temperature dependence of Cs volatilisation from Fukushima adsorbents	Dimitri Pletser  Imperial College
12:50	A DISTINCTIVELY academic career	Dr. Claire Corkhill  University of Sheffield
13:10	Lunch & Poster Session	
<b>Session 2</b> <i>Chairs: Prof. Colin Boxall, Lancaster University &amp; Prof. David Read, University of Surrey</i>		
14:10	Innovation at Sellafield, the new Integrated Innovation Teams	Dr Stephen Hepworth  Sellafield Ltd
14:50	Microstructural Characterisation of AGR Cladding Materials	Chiara Barcellini  The University of Manchester
15:10	Direct Mass Analysis of Water Absorption onto Cerium, Thorium and Uranium oxides	Dr. Dominic Laventine  University of Lancaster
15:30	Refreshments & Poster Session	
16:00	The PhD after-life: Working for the supply chain	Dr. Carlos De La Fontaine  TUV-SUD Nuclear Technologies

16:20	<b>Switching on Ion Exchange in Metal Germanates</b>	Ryan George University of Birmingham
16:40	<b>Characterization of cement based materials through Atomic Force Microscopy</b>	Luca Rizzo University of Strathclyde
17:00	Break	
18:30	Drinks & Canapés Reception	National Railway Museum
19:30	Dinner	National Railway Museum 

Thursday 6 <sup>th</sup> April		
Time	Title	Presenter
08:30	Registration / Poster Board set up / Refreshments	
09:00	Introduction and Housekeeping	Prof. Michael Fairweather University of Leeds
<b>Session 3</b> Chairs: Dr. Joe Hriljac, University of Birmingham & Prof. Michael Fairweather, University of Leeds		
09:10	<b>Hanford: The Creation and Remediation of the Legacy</b>	Dr Albert Kruger DOE Office of River Protection
09:50	Raman probe development : An investigation into active sludge components	Kate Wyness University of Bristol
10:10	<b>Robotic vision for 3D modelling, detection and recognition of nuclear waste objects and scenes</b>	Henry Zhao University of Birmingham
10:30	<b>Nanoparticle Technology in Waste Disposal and Remediation Systems</b>	Eleonora Cali Imperial College
10:50	Refreshments & Poster Session	
11:20	<b>Geological Disposal: Deep down, you know it makes sense</b>	Dr. Amy Shelton NDA

11:40	Glass-ceramics for Pu disposition - where are we now?	Steph Thornber University of Sheffield
12:00	Quartz crystal microbalance as a tool to measure complex suspension rheology	Andre Botha University of Leeds
12:20	The Behaviour of Spent Nuclear Fuel in Wet Storage	Elizabeth Howett University of Lancaster
12:40 Lunch and Poster Session		
Session 4 Chairs: Prof. Rebecca Lunn, University of Strathclyde & Professor Tom Scott, University of Bristol		
13:40	Transitioning of Spent AGR Fuel from Wet to Dry Storage	James Goode University of Leeds
14:00	Smart cements for chloride / moisture sensing in nuclear concrete assets	Lorena Biondi University of Strathclyde
14:20	Heat Treatments of Cl-- and Water-contaminated PuO2 at NNL	Sophie Sutherland-Harper The University of Manchester
14:40	Online synchrotron-based uranium corrosion experiments	Haris Parasevoulakos University of Bristol
15:00 Refreshments & Poster Session		
15:30	The effects of colloidal silica based grouts on Sr and Cs speciation – present and future research	Dr. Pieter Bots University of Strathclyde
15:50	Gas generation from water on the surface of plutonium dioxide	Dr. Luke Jones The University of Manchester
16:10	Adventures in Actinide Science	Dr Tamara Griffiths NNL
16:30	An update on Impact	Prof Neil Hyatt University of Sheffield
16:50	NDA PhD Student Awards	 Nuclear Decommissioning Authority Deborah Ward, NDA
17:00	Closing remarks	Prof. Michael Fairweather University of Leeds

**Theme 1:    AGR, Magnox and Exotic Spent Fuels**

**Theme 2:    PuO<sub>2</sub> and Fuel Residues**

**Theme 3:    Legacy Ponds and Silo Wastes**

**Theme 4:    Structural Integrity**

**Keynote**

**Employability**

**Cross-cutting themes**

Shaded name = eligible for the £250 prize for best oral presentation given by a DISTINCTIVE PhD student.

**All presentations will be made available to download from our consortium website:**

**<http://distinctiveconsortium.org/category/events/annual-meeting-events/>**

## Posters

The following posters will be presented throughout the event:

Poster Number	Researcher	Poster title	University
1	Sophie Rennie	The Effect of Crystal Orientation on the Radiolytic Dissolution of UO <sub>2</sub>	Bristol
2	Claudia Gasparrini	Uranium carbide oxidation: an <i>in situ</i> high temperature environmental SEM study	Imperial
3	Elizabeth Howett	The Behaviour of Spent Nuclear Fuel in Wet Storage	Lancaster
4	James Goode	Transitioning of Spent AGR Fuel from Wet to Dry Storage	Leeds
5	Chiara Barcellini	Grain Boundary Damage Mechanism in AGR Cladding Material under Irradiations	Manchester
6	Andrea Paulillo	Assessing the impact of nuclear waste management in the UK	UCL
7	Nathan Palmer	Atomistic simulations of helium incorporation in PuO <sub>2</sub> and surface modelling.	Birmingham
8	Rashed Sarwar	Real-time fast-neutron plutonium assay for storage and ageing applications.	Lancaster
9	Dr Dominic Laventine	Direct Mass Analysis of Water Absorption onto Cerium, Thorium and Uranium oxides	Lancaster
10	Dr Bengt Tegner	Water Adsorption on Actinide Oxide Surfaces	Manchester
11	Marisa Smith	Importance of Secondary Electrons in Water Irradiation	Manchester
12	Sophie Sutherland-Harper	PuO <sub>2</sub> studies at NNL	Manchester
13	Dr Shi-Kuan Sun	Ceramic and Glass-Ceramic Wasteforms for Actinide Disposition	Sheffield
14	Steph Thornber	Zirconolite glass-ceramics for the disposition for Pu-residues.	Sheffield
15	Antonia Yorkshire	Understanding radionuclide interactions with cementitious materials for radioactive waste management	Sheffield
16	Dr Thomas Donoclift	A Catalytic Approach to H <sub>2</sub> and O <sub>2</sub> Recombination on PuO <sub>2</sub> and PuO <sub>2</sub> Surrogates	Manchester

17	Ryan George	Switching on Ion Exchange in Metal Germanates.	Birmingham
18	George Day	Thermal conversion of Sr-loaded IONSIV	Birmingham
19	Dr Evin (Tzu-Yu) Chen	Metal substitution and Cs/Sr Ion Exchange in Sn-silicate Umbites	Birmingham
20	Kate Wyness	Raman probe development: Sample preparation for uranium-oxides synthesis	Bristol
21	Dr Paul Fossati	Atomistic modelling of glass/crystal interfaces	Imperial
22	Dimitri Pletser	Low Temperature Immobilisation of Spent Adsorbents from Fukushima.	Imperial
23	Eleonora Cali	Improved Uranium Removal from Effluents using Binding Superparamagnetic Nanoparticles	Imperial
24	Olivia Lynes	ab initio Molecular Dynamics Studies of Hydrated Hydroxides of Strontium and other Alkali Earth Metals	Lancaster
25	Alastair Tonge	Novel Characterisation of Flocculated Dispersions using Acoustic Backscatter Systems	Leeds
26	Michael Johnson	An x-ray tomography study of the evolution of bubble populations in nuclear legacy waste	Leeds
27	Dr Derrick Njobuenwu	Simulation of dense suspension in pipe flows with homogeneous stationary sediment bed	Leeds
28	Dr Hugh Rice	In-line rheometry and flow characterisation of dense slurries in pipe flow using acoustic methods	Leeds
29	Andre Botha	Quartz crystal microbalance as a tool to measure complex suspension rheology	Leeds
30	Keith Schou	Application and theory of shear enhanced ultrafiltration with calcite suspension	Loughborough
31	Olusola Ayoola	Feasibility of Ultrasonic Spectroscopy for Use In-Situ Sludge Characterisation	Manchester
32	Luke Boast	Thermal treatment of plutonium contaminated materials	Sheffield
33	Henry Cheng Zhao	Robotic vision for 3D modeling, detection and recognition of nuclear waste objects and scenes	Birmingham
34	Toby Lord	Laser Induced Breakdown Spectroscopy for Contamination Detection in Concrete.	Leeds



35	Ryan Kavanagh	Investigating the effects of ionizing radiation on calcium silicate hydrates using first-principles calculations	QUB
36	Tom Mullan	Production of biogenic phosphate minerals and applications for the remediation of radionuclides	Strathclyde
37	Dr Matteo Pedrotti	Use of colloidal silica grout for ground barriers in decommissioning	Strathclyde
38	Dr Pieter Bots	The effects of colloidal silica based grouts on Sr and Cs speciation – present and future research	Strathclyde
39	Luca Rizzo	Characterization of cement based materials through Atomic Force Microscopy	Strathclyde
40	Mel O'Leary	Irradiated Sludges	QUB

Shaded name = eligible for the £250 prize for the best poster presentation given by a **DISTINCTIVE PhD** student.

## Attendee List

First Name	Surname	Organisation
Anna	Adamska	Sellafield
Mike	Angus	NNL
Olusola	Ayoola	University of Manchester
Sarah	Ashwood	EPSRC
Antonios	Banos	University of Bristol
Chiara	Barcellini	University of Manchester
Martyn	Barnes	Sellafield Ltd
Lorena	Biondi	University of Strathclyde, CEE
Richard	Blackham	Sellafield Ltd
Luke	Boast	University of Sheffield
Gary	Bolton	National Nuclear Laboratory
Andre	Botha	University of Leeds
Pieter	Bots	University of Strathclyde
Dominic	Box	Fraser-Nash Consultancy
Gunnar	Buckau	EC - JRC
Ed	Butcher	National Nuclear Laboratory
Eleonora	Cali'	Imperial College London
Colin	Campbell	Environment Agency
Thomas	Carey	NNL
Tzu-Yu	Chen	University of Birmingham
Sandra	Clarke	Sellafield Ltd
Jonathan	Collard	University of Manchester
Paul	Cook	Sellafield
Andrew	Cooney	Sellafield Ltd
Duncan	Coppersthwaite	National Nuclear Laboratory
Claire	Corkhill	University of Sheffield
Matthew	Cotton	University of York
Robin	Cowley	Sellafield Ltd.
James Ed	Darnbrough	University of Bristol
George	Day	University of Birmingham
Carlos	De La Fontaine	TUV SUD Nuclear Technologies
Thomas	Donoclift	University of Manchester
Koulis	Efkarpidis	Sellafield Ltd
Michael	Fairweather	University of Leeds
Paul	Fossati	Imperial College London
Ryan	George	University of Birmingham
James	Goode	University of Leeds
James	Graham	National Nuclear Laboratory
Jeffrey	Griffin	Savannah River National Laboratory
Tamara	Griffiths	NNL
Antonio	Guida	Amec Foster Wheeler

David	Harbottle	University of Leeds
Mike	Harrison	NNL
Bruce	Hanson	University of Leeds
Steve	Hepworth	Sellafield Ltd
Joanne	Hill	Hydrock NMCL
Jeff	Hobbs	Sellafield Ltd
Elizabeth	Howett	Lancaster University
Joe	Hriljac	University of Birmingham
Timothy	Hunter	University of Leeds
Neil	Hyatt	University of Sheffield
Mike	James	Sellafield Ltd
Michael	Johnson	University of Leeds
Luke	Jones	University of Manchester
John	Jowsey	Sellafield Ltd
Laurie	Judd	Longnecker & Associates
Ryan	Kavanagh	Queens University Belfast
Simon	Kellet	Sellafield Ltd
Albert	Kruger	US Department of Energy
Dominic	Laventine	Lancaster University
Laura	Leay	Dalton Cumbrian Facility   The University of Manchester
Bryony	Livesey	Costain
Toby	Lord	University of Leeds
Rebecca	Lunn	University of Strathclyde
Olivia	Lynes	Lancaster University
Graham	Mackay	National Nuclear Laboratory
Ewan	Maddrell	National Nuclear Laboratory
Mithila	Manage	National Nuclear Laboratory
Darryl	Messer	University of Manchester
Sean	Morgan	Sellafield Ltd
Thomas	Mullan	University of Strathclyde
Derrick	Njobuenwu	University of Leeds
Mel	O'Leary	Queen's University Belfast
Peter	Orr	EA
Scott	Owens	Frazer-Nash / Imperial College
Cristiano	Padovani	AMEC-FW
Stephen	Palethorpe	National Nuclear Laboratory
Nathan	Palmer	The University of Birmingham
Haris	Paraskevoulakos	University of Bristol
Hannah	Paterson	Sellafield Ltd
Andrea	Paulillo	University College London
Matteo	Pedrotti	University of Strathclyde
Ian	Pegg	The Catholic University of America
Marcus	Perry	University of Strathclyde

Simon	Pimblott	The University of Manchester
Dimitri	Pletser	Imperial College London
Geoff	Randall	Sellafield Ltd
David	Read	University of Surrey
Sophie	Rennie	University of Bristol
Joanna	Renshaw	University of Strathclyde
Hugh	Rice	University of Leeds
Rodrigo	Rimando	U.S. Department of Energy
Luca	Rizzo	University Of Strathclyde
Rebecca	Robbins	IAEA
Bill	Rogerson	Sellafield Ltd.
Mary	Ryan	Imperial College London
Rashed	Sarwar	Lancaster Univeristy
Keith	Schou	Loughborough University
Amy	Shelton	Radioactive Waste Management
Rick	Short	Nuclear Decommissioning Authority
Marisa	Smith	University of Manchester
Jon	Squire	Sellafield Ltd
Helen	Steele	Sellafield Ltd
Rob	Stephen	Sellafield Ltd
Shikuan	Sun	University of Sheffield
Jake	Surman	Environment Agency
Sophie	Sutherland-Harper	University of Manchester
Frank	Taylor	LLW Repository Ltd
Bengt	Tegner	The University of Manchester
Paul	Thompson	AWE
Stephanie	Thornber	University of Sheffield
Alastair	Tonge	University of Leeds
Lois	Tovey	University of Leeds
Luc	Vandeperre	Imperial College London
John	Vienna	Pacific Northwest National Laboratory
Steve	Walters	National Nuclear Laboratory
Simon	Woodbury	National Nuclear Laboratory
Kate	Wyness	University of Bristol
Antonia	Yorkshire	University of Sheffield
Cheng	Zhao	University of Birmingham

## Master Project List

Theme	Title	Leading Institution	Researcher	Type
1	An Investigation of Wasteform Evolution During Wet-recovery and Drying of SNF	Bristol	Dr. James E Darnbrough Dr. Leila Costelle	PDRA
1	UO <sub>2</sub> Surface Reactivity and Alteration – a Fundamental Study of Photocatalytic and Structural Effects Related to Long Term Storage of SNF	Bristol	Sophie Rennie	PhD
1	Options for Exotic Carbide Fuels	Imperial	Claudia Gasparini	PhD
1	The Behaviour of Used Nuclear Fuel in Wet Storage	Lancaster	Elizabeth Howett	PhD
1	Determination of Optimum Drying Conditions for AGR fuels	Leeds	James Goode	PhD
1	Grain Boundary Damage Mechanisms in Strained AGR Cladding Under Irradiation	Manchester	Chiara Barcellini	PhD
1	A Life Cycle Approach as a Decision Tool for Nuclear Waste Management and Decommissioning of Existing and Future Plants	UCL	Andrea Paulillo	PhD
2	Computational Modelling of PuO <sub>2</sub> Ageing and Fuel Residues	Birmingham	Nathan Palmer	PhD
2	In-situ Characterisation of Heavily-Contaminated Plutonium Finishing Environments	Lancaster	-	PhD
2	Real-time Fast Neutron Plutonium Assay for Plutonium Storage and Ageing Applications	Lancaster	Rashed Sarwar	PhD
2	Understanding the Interfacial Interactions of Plutonium Dioxide with Water	Lancaster	Dr. Dominic Laventine	PDRA
2	Investigation of Anomalous Hydrogen Production from Water Adsorbed on Oxides	Manchester	Jamie Southworth	PhD
2	Modelling the Surface Chemistry of PuO <sub>2</sub> at the Molecular Level	Manchester	Dr. Bengt Tegner	PDRA
2	Simulation of Low-energy Electron Radiolysis of Water Adsorbed on Oxides	Manchester	Marisa Smith	PhD
2	Understanding Surface Species and Interactions Between Adsorbed Chloride and Water on Stored PuO <sub>2</sub>	Manchester	Sophie Sutherland-Harper	PhD
2	Understanding the Interfacial Interactions of Plutonium Dioxide with Water	Manchester	Dr. Luke Jones Dr Thomas Donoclift	PDRA
2	Ceramic Materials for Actinide Disposition	Sheffield	Dr. Shi-Kuan Sun	PDRA

2	Development of Glass-ceramics for Pu Disposition using Hot Isostatic Pressing	Sheffield	Stephanie Thornber	PhD
2	Understanding Actinide Sorption and Binding to Cement Materials for Radioactive Waste Management	Sheffield	Antonia Yorkshire	PhD
2	The Interaction of Water with PuO <sub>2</sub> Surfaces	UCL	Joseph Wellington	PhD
3	New Ion Exchange Materials For Effluent Clean-up	Birmingham	Ryan George	PhD
3	Novel Ceramic Wasteforms for Cs and Sr Encapsulation	Birmingham	George Day	PhD
3	Novel Ion Exchange Materials	Birmingham	Dr. Evin (Tzu-Yu) Chen	PDRA
3	Corrosion of Uranium in Water and Hydrogen	Bristol	Antonis Banos	PhD
3	Development of Raman Spectroscopy Techniques for the Remote Analysis of Nuclear Wastes in Storage	Bristol	Kate Wyness	PhD
3	The Evolution of Grouted Waste Forms Containing Uranium	Bristol	Haris Paraskevoulakos	PhD
3	Durability of Heterogeneous ILW Glass/Ceramic Wasteforms from Complex Wastestreams	Imperial	Dr. Paul Fossati	PDRA
3	Glass Composite Materials for Fukushima ILW Immobilisation	Imperial	Dimitri Pletser	PhD
3	Glass Composite Materials for Sellafield LP&S ILW Immobilisation	Imperial	Charles Hutchison	PhD
3	Magnetic Nanoparticles for Waste Separation or Sequestration	Imperial	Eleonora Cali	PhD
3	Computational Simulations of Storage Pond Sludge Disturbance	Lancaster	Olivia Lynes	PhD
3	Characterisation of Flocculated Waste Suspensions with Acoustic Backscatter	Leeds	Alastair Tonge	PhD
3	Gas Retention and Release from Nuclear Legacy Waste	Leeds	Michael Johnson	PhD
3	Measurement and Modelling of Sludge Mobilisation and Transport	Leeds	Dr. Derrick Njobuenwu/Dr. Hugh Rice	PDRA
3	The Development of Characterisation Techniques for Intermediate Level Waste Sludges	Leeds	Andre Botha	PhD
3	Enhanced Shear Micro- and Ultra-Filtration Without Recycle Pumping	Loughborough	Keith Schou	PhD

3	Autonomous Systems for Nuclear Decommissioning in Extreme Radiation Environments	Manchester	Olusola Ayoola	PhD
3	Irradiated Sludges - Experimental	QUB	Mel O'Leary	PhD
3	Modelling Hydrogen Generation from Radioactive Sludges	QUB	Conrad Johnston	PhD
3	Thermal Treatment of PCM and ILW	Sheffield	Luke Boast	PhD
3	The Interaction of Brucite Surfaces with Uranium and its Fission Products	UCL	Eszter Makkos	PhD
4	Production of Real-time Segmented as-built CAD Models for the Planning and Execution of Remote and Human Intervention Tasks	Birmingham	Henry (Cheng) Zhao	PhD
4	The Impact of Recycled Concrete Fines on the Engineering Performance of Cementitious Infill	Leeds	Toby Lord	PhD
4	Simulating Radiation Damage in Cement	QUB	Ryan Kavanagh	PhD
4	Crack Sealing and Water Transport	Strathclyde	Riccardo Maddalena	PhD
4	Development of Novel, Low Cost Biomineral Permeable Reactive Barriers for Radionuclide Remediation	Strathclyde	Tom Mullan	PhD
4	In-situ Ground Contaminant Containment (Physical barrier)	Strathclyde	Christopher Wong	PhD
4	In-situ Ground Contaminant Containment (Physical barrier)	Strathclyde	Dr. Matteo Pedrotti	PDRA
4	Integrated Sensors for Infrastructure	Strathclyde	-	PhD
4	Nano-cracking of Cement Phases: Reactivity and Dissolution	Strathclyde	Luca Rizzo	PhD
4	Smart cements for chloride and moisture sensing in nuclear concrete assets	Strathclyde	Lorena Biondi	PhD
4	Impacts of colloidal silica based grout injection on geo-mechanical and geochemical properties of soil and radioactive wastes	Strathclyde	Dr Pieter Bots	PDRA

# Abstracts

Full project summaries can be downloaded from our consortium website:

<http://distinctiveconsortium.org/category/events/annual-meeting-events/>

The following pages contain the abstracts (in the order of the master project list) from these summaries.



# Theme 1 - AGR, Magnox and Exotic Spent Fuels

## Radiation Driven Reactions at the Surface of Uranium Dioxide

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### Abstract

In order to ensure resilient, long-term storage for nuclear material it is critical to have a thorough understanding of the reactions occurring at the surface of stored uranium oxides. This project aims to explore this topic in further detail through studying radiation induced oxidative dissolution of uranium dioxide thin films. Building on our previously developed experimental technique, we have expanded upon our initial measurements to investigate the effect of crystal orientation on the dissolution of UO<sub>2</sub>. These measurements were conducted on the I07 beamline at the Diamond Light Source in March 2016 and a full analysis of the experimental results is currently underway. However, preliminary analysis has shown crystal orientation to significantly affect the rate of UO<sub>2</sub> dissolution, with the [111] film appearing far more resilient to radiation induced corrosion than either the [001] or [110] orientations.

## Uranium carbide and zirconium carbide oxidation: an *in situ* high temperature environmental SEM study

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<sup>5</sup> Commissariat à l'Energie Atomique, CEA, CADARACHE, France

### Abstract

Oxidation of depleted uranium carbide (UC) was investigated via experiments on sintered fragments oxidised in air in a furnace and *in situ* with a state-of-the-art technique: High temperature Environmental Scanning Electron Microscopy (HT-ESEM). This technique revealed the morphological changes occurring during the transformation between UC to UO<sub>2</sub> and UO<sub>2</sub> to U<sub>3</sub>O<sub>8</sub>. Experiments performed in an atmosphere of 10 Pa of oxygen and in a furnace in air from 873 – 1173 K revealed the influence of temperature in the final oxide product characteristics: oxide morphology, conversion and specific surface area (SSA). Samples oxidised at T > 873 K showed partial sintering of the oxide which acts as a diffusion barrier and limits the occurrence of UO<sub>2</sub> to U<sub>3</sub>O<sub>8</sub> to cracked surfaces. Experiments performed from 723 – 848 K in an atmosphere of 10 to 100 Pa O<sub>2</sub> revealed the key role

of crack propagation, monitored with an image analysis technique, in the oxidation and the self-ignition process of UC.

Prior to UC active work, zirconium carbide (ZrC) oxidation was investigated. Oxidation performed from 1073 – 1473 K in air and oxygen atmosphere revealed the interface in between the carbide and the oxide to be comprised of an amorphous carbon matrix with nanocrystals of zirconia embedded in it. *In situ* analysis performed on ZrC with an HT-ESEM was used to examine the influence of crack propagation on the Maltese Cross shape development of the oxide.

## **The Behaviour of Spent Nuclear Fuel in Wet Storage**

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### **Abstract**

The project aims to investigate the behaviour of Spent Nuclear Fuel (SNF) in interim storage, specifically AGR fuel in storage ponds at Sellafield. Corrosion behaviour will be studied in order to assess the validity of extended storage periods. Studies will be carried out on UO<sub>2</sub>, SIMFUEL and AGR cladding samples separately and in binary systems. Once these initial experiments have been completed, analogous experiments will be carried out on real spent AGR fuel. Studies have been carried out on the corrosion behaviour of sensitised and unsensitised stainless steel cladding samples in simulant pond water with varying temperature, chloride concentration, pH and hydrogen peroxide concentration.

## **Transitioning of Spent AGR Fuel from Wet to Dry Storage**

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### **Abstract**

UK strategy relating to the treatment of spent nuclear fuel (SNF) has recently changed from reprocessing as part of a closed fuel cycle to direct disposal into a geological disposal facility (GDF). Since a GDF is not expected to be available until 2075 interim storage of fuel will be required. The use of interim dry storage has been mooted however, little research has been carried out into the effects of drying stainless steel (SS) clad spent nuclear fuel and this PhD is intended to begin work in this area.

Preliminary work has been carried out on simulant aluminium clad fuels as part of an MSc project which has been used to evaluate some initial experimental methods, however going forward work will concentrate on SS clad fuels. The initial phase of this work will be the development of a method for producing and characterising samples representative of SNF that has been stored for 20 years in caustic conditions (Sellafield's current strategy).

## Grain Boundary Damage Mechanism in AGR Claddings under Irradiation

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### Abstract

The safe interim storage of AGR irradiated fuel in water ponds requires a detailed knowledge of the corrosion behaviour of the fuel cladding material. The systematic study of corrosion relies on the availability of sensitised 20Cr25Ni Nb-stabilised stainless steel, ideally with a microstructure close to that produced by neutron irradiation but less radioactive. The aim of this PhD project is the production of sensitised specimens using an intense beam of protons, and to develop thereupon an understanding of the mechanisms driving the radiation-induced segregation of chemical species in the vicinity of grain boundaries. Analytical electron microscopy will be used to investigate the damaged structures resulting from the irradiation experiments, in order to compare them with those of claddings removed from the reactor core. A profound knowledge of the behaviour of 20Cr25Ni Nb-stabilised stainless steel under irradiation, however, starts with the characterisation in the pre-irradiation conditions (cold-rolled and annealed), and to determine the optimum conditions for the ion irradiation experiments.

## Theme 2 – PuO<sub>2</sub> and Fuel Residues

### Atomistic simulations of helium incorporation in PuO<sub>2</sub> and surface modelling

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<sup>2</sup> Department of Chemistry, University of Bath, Claverton Down, Bath, BA2 7AY, United Kingdom

### Abstract

Static lattice simulations have been performed using robust and accurate potentials to gain insight into properties of PuO<sub>2</sub>. Using the GULP code [1], the potentials by Read et al. (2014) [2] and Grimes et al. (1990) [3] have been used to model helium atom incorporation in pure and defective PuO<sub>2</sub> lattice. The results show that helium gas incorporation in octahedral sites is endothermic. In addition, they show that plutonium vacancies are the preferred trapping sites, in agreement with other studies for UO<sub>2</sub> [4-6].

In addition, migration energy barriers between oxygen ions are predicted to be appreciably high at around 4 eV. Pure PuO<sub>2</sub> surfaces have been modelled using the METADISE program [7]. The simulations show that the (111) surface is the most energetically stable surface, followed by the (221) and (331) then the (110) surface. The (221) and (331) surfaces are relatively stable surfaces, exhibiting complicated faceted structures, which would be predicted to appear in the morphology of PuO<sub>2</sub>.

## Real-time fast-neutron plutonium assay for storage and ageing applications

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### Abstract

Nuclear safeguards is the discipline that ensures that civilian nuclear installations are not being misused to pursue weapons and that associated materials are not diverted to illegal usage. The well-established techniques for verifying this include passive/active radiation monitors using one of several detection methods including gamma-ray tomography, neutron coincidence counters using He-3 detectors and the Cherenkov viewing device etc. Each method has its benefits but individual limitations too. Additionally, the development of advanced fuel-cycle material, such as mixed-oxide (MOX) fuel has indicated further limitations of existing safeguard techniques due to presence, for example, of multiple actinides acting like neutron sources having signatures comparable to plutonium. A solution to this can be achieved by using liquid scintillation detectors (LSD) for fast neutron multiplicity analysis or coincidence counting. These detectors are sensitive to both fast neutrons and gamma radiation. The primary advantage of using such detectors is that they enable an extremely short gate time (three orders of magnitude lower than He-3 detectors) to be used, allowing reduced accidental coincidences and thus being able to detect higher orders of multiplicity.

This studentship is supported by the National Nuclear Laboratory as part of the EPSRC DISTINCTIVE consortium along with Lancaster University with the aim of (i) identifying the multiplicity distribution of a sample in real-time using liquid scintillation detectors to discriminate between different isotopes and (ii) exploiting ( $\alpha$ , n) reactions to distinguish the uncorrelated events from the correlated neutrons. This report primarily focuses on the design and development of a real-time system capable of the rapid data processing needed to this end. This is required due to the very short pulse length (in the order of 50-70 ns) originating from LSD when radiation is detected. Moreover, considering practical geometry of an experimental setup (i.e. several rings of detectors surrounding a source within a radius of 50-100cm) and the high speed at which fast neutrons travel, it is expected that fission neutrons will theoretically arrive at the detector after within 20 to 40 ns of the fission event taking place. Hence it is imperative that the system is capable of sampling the detector response at 200 MHz or higher. This feat was achieved utilizing a platform based on System-on-Chip Field-Programmable Gate Array (SoC-FPGA) which combines a dual-core Cortex-A9 embedded core with a FPGA fabric using a high-bandwidth interconnecting link. The system is capable of constructing neutron coincidence distributions in real-time by sampling up to 16 detectors every 5 ns over a user-defined gate-width or coincidence window. This distribution can then be unfolded to carry out neutron multiplicity analysis.

## The Behaviour of Used Nuclear Fuel in Wet Storage

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### Abstract

Plutonium oxide (PuO<sub>2</sub>) is one of the most highly radioactive components of nuclear fuel waste streams and its storage poses particular challenges due to the high temperatures produced by its decay and the production of gases (particularly H<sub>2</sub> and steam). Its high radiotoxicity necessitates the use of analogues, such as ceria and urania, to allow the comprehensive study of its interaction with water under storage conditions. We have developed a method which enables direct gravimetric measurement of water adsorption onto CeO<sub>2</sub> thin films with masses in the microgram region. Porous CeO<sub>2</sub> films were fabricated from a surfactant based precursor solution. The absorption of water onto the CeO<sub>2</sub> coating at different relative humidities was studied in a closed reactor. Quartz Crystal Microbalance (QCM) gravimetry was used as a signal transducer, as changes in crystal resonant frequency due to absorbed mass are directly and linearly related to mass changes occurring at the

crystal surface. Using this method, we have determined the enthalpy of absorption of water onto CeO<sub>2</sub> to be 49.7 kJmol<sup>-1</sup> at 75°C, and onto U<sub>3</sub>O<sub>8</sub> to be 54.1 kJmol<sup>-1</sup> at 75°C, 11 and 15 kJmol<sup>-1</sup> greater than the enthalpy of evaporation, respectively. These enthalpy is within the range predicted for the absorption of water onto PuO<sub>2</sub>, indicating this method allows for investigation of water absorption using microgram samples.

## Water Adsorption on Reduced Actinide Oxide Surfaces

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### Abstract

The interactions between water and the actinide oxides UO<sub>2</sub> and PuO<sub>2</sub> are important when considering the long-term storage of spent nuclear fuel. However, experimental studies in this area are severely limited by plutonium's intense radioactivity, and hence we have recently begun to investigate these interactions computationally. In this summary we report the results of first principles calculations of the interaction of water with UO<sub>2</sub> surfaces. Strongly-correlated effects are taken into account using a Hubbard corrected potential, which enables us to perform efficacious plane-wave density functional calculations of extended systems. In particular, we compare results of water adsorption on reduced UO<sub>2</sub> {111}, {110} and {100} with the corresponding results on CeO<sub>2</sub>, focusing on the energetics and structural properties of molecular versus dissociative adsorption, on or near an oxygen vacancy.

## Importance of Secondary Electrons in Water Irradiation

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### Abstract

Monte Carlo simulation of the track structure of low energy electrons in water has been performed using experimental cross-sections for the collision processes in amorphous solid water and gaseous water scaled to liquid density. There are significant differences between the predictions of the two calculations for the number of interactions and the thermalisation distance. These differences between the two sets of simulations clearly demonstrate that collective effects due to condensation, i.e. interaction between molecules, are important. The use of gas phase cross-section to simulate energy loss processes of low energy electrons in condensed water, where in bulk or adsorbed onto surfaces, is inappropriate and an erroneous point to start modelling radiation chemical kinetics.

## Methodology for the Heat Treatments of Chloride- and Water-contaminated PuO<sub>2</sub> at NNL

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### Abstract

Heat treatment of chloride-contaminated PuO<sub>2</sub> powders has been carried out at the National Nuclear Laboratory, Cumbria using a furnace in an air glove box to volatilise the chloride species. Untreated PuO<sub>2</sub> and

PuO<sub>2</sub> heat treated to a range of temperatures have been analysed by X-Ray Diffraction. Water-adsorption experiments on PuO<sub>2</sub> powders have also been undertaken in a sealed vessel by heating the powders and measuring the pressure as they cool in the presence of water. The following paper is a review of the technical methodology used to perform these experiments and analyses.

## **Gas Generation from Water on the Surface of Plutonium Dioxide**

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<sup>4</sup> Sellafield Sites Ltd, Sellafield, Seascale, Cumbria CA20 1PG

### **Abstract**

The long term storage of separated plutonium dioxide (PuO<sub>2</sub>) in sealed canisters requires an understanding of the processes occurring within them. There are several processes with the potential to pressurise these canisters. One of these processes is the radiolysis of adsorbed water leading to the formation of hydrogen (H<sub>2</sub>) and oxygen (O<sub>2</sub>). A review of previous work investigating H<sub>2</sub> production from the interaction of water and PuO<sub>2</sub> and discussion of the results is undertaken. An outline of experiments currently ongoing at the National Nuclear Laboratory is also discussed.

## **A Catalytic Approach to H<sub>2</sub> and O<sub>2</sub> Recombination on PuO<sub>2</sub> and PuO<sub>2</sub> Surrogates**

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### **Abstract**

A typical plutonium dioxide storage package contains very little water, and as a result, package pressurisation due to alpha radiolysis of water is an anomalous phenomenon, observed in only a few “out-of-specification” packages. Previous measurements of hydrogen production from the surface of dry PuO<sub>2</sub> suggest that package pressurisation should be more common. As this is contrary to what is observed at Sellafield, it must be concluded that some removal mechanism is taking place. This work aims to measure the rate of recombination of H<sub>2</sub> and O<sub>2</sub> over plutonium dioxide. By taking a catalytic approach it is hoped that experimental measurements can provide data to reduce some of the pessimisms built into the Compliance Index model and give insight to the chemical mechanism of recombination taking place on the surface of plutonium dioxide.

## Ceramic and glass-ceramic wasteforms for actinide disposition

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### Abstract

Pyrochlore ceramic and zirconolite glass-ceramic wasteforms are the potential host phases for actinide disposition. Composition – processing – structure relations in a series of wasteforms (uranium betafite, cerium pyrochlore and zirconolite glass ceramic) was investigated in this work. Fully dense uranium betafite ceramic was prepared by sintering of the oxide precursors ( $\text{CaTiO}_3$ ,  $\text{TiO}_2$ ,  $\text{ZrO}_2$  and  $\text{U}_3\text{O}_8$ ) at 1320°C in flowing  $\text{N}_2$ . The betafite phase in high yield (> 85%) with uranium oxidation state of  $4.2 \pm 0.1$  was found in the resulted ceramics and all  $\text{U}_3\text{O}_8$  was incorporated into the ceramic. As a Pu surrogate, cerium pyrochlore with nearly single phase was obtained and the crystal structure (Space group  $\text{Fd-}3\text{m}$ ,  $a = 10.1469 \text{ \AA}$ ) was determined from neutron diffraction data. After heavy ion irradiation on zirconolite glass-ceramic, the crystallinity of zirconolite phase decreased in the damaged surface layer, and the swelling feature of glass phase was observed along the zirconolite grain boundary. The morphology evolution during the dissolution at 90°C in water showed the glass dissolved first and the depth difference between zirconolite and glass phase after 28 days was around 2.0  $\mu\text{m}$ .

## Ce Incorporation in Zirconolite Glass-Ceramics

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### Abstract

The UK has 140 tonnes of separated civil  $\text{PuO}_2$  stored at the Sellafield site. The strategy for dealing with this stockpile is to fabricate MOx fuel, however a significant fraction has been identified as higher activity waste requiring immobilisation into stable wasteforms. In addition, Pu-residues are a classification of Pu-bearing higher activity wastes that range in Pu concentration, composition and physical form, thus requiring flexible wasteforms for immobilisation. This project investigates zirconolite glass-ceramics as future wasteforms for impure streams of these Pu wastes. Consolidated by hot isostatic pressing, Ce was targeted for incorporation on either the  $\text{Ca}^{2+}$  and / or  $\text{Zr}^{4+}$  sites as either  $\text{Ce}^{3+}$  or  $\text{Ce}^{4+}$ . The formation of a Ce-bearing perovskite phase was the result of Ce reduction when targeting Zr substitution, and has been discussed on the account of ionic radii arguments.

## Understanding radionuclide interactions with cementitious materials for radioactive waste management

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### Abstract

In the UK, the current method for immobilisation of intermediate level waste (ILW) streams is encapsulation in Portland cement grouts. Cement hydrate phases, which form during the hydration of cement clinker and pozzolanic additives, are capable of binding radionuclides. One of the key issues associated with cement encapsulation of ILW is the uncertainty of radionuclide fate within the cementitious matrix over the long timescales and varying local conditions of extended interim storage and geological disposal. Discussed here are

the syntheses of singular cement hydrate minerals including Calcium-Silicate-Hydrate (CSH), hydrotalcite ( $\text{Mg}_6\text{Al}_2\text{CO}_3(\text{OH})_{16}\cdot 4\text{H}_2\text{O}$ ) and ettringite ( $\text{Ca}_6(\text{Al}_2\text{O}_6)(\text{SO}_4)_3\cdot 32\text{H}_2\text{O}$ ). These minerals have been exposed to solutions of U, Tc, Ce and Pu to develop an understanding of the mechanisms of uptake and incorporation into simplified systems for those Portland cement systems used for ILW. These experiments form the focus of longer term sorption / de-sorption studies to observe long-term radionuclide incorporation cements. The results of uranium incorporation in CSH are discussed in detail, as are the results from experiments performed at the National Nuclear Laboratory, where particles of  $\text{PuO}_2$  were encapsulated in Portland cement blends that are used in the encapsulation of ILW, including Plutonium Containing Material (PCM).

## Theme 3 – Silo Ponds and Legacy Wastes

### Switching on Ion Exchange in Metal Germanates

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#### Abstract

The synthesis of materials for ion exchange and subsequent disposal of radionuclides will be explored in this work. The successful synthesis of the germanium form of the natural mineral umbite ( $\text{K}_2\text{ZrSi}_3\text{O}_9\cdot \text{H}_2\text{O}$ ) and subsequent ion exchange results shows both caesium and strontium uptake by Ge-Zr-umbite ( $\text{K}_2\text{ZrGe}_3\text{O}_9\cdot \text{H}_2\text{O}$ ) but at low levels. This is substantially improved by chemical modification of the umbite structure with the incorporation of up to 30% Nb for Zr at the octahedral site. Further doping results in mixed phases with the formation of umbite and another interesting pharmacosiderite phase ( $\text{HK}_3\text{Ge}_7\text{O}_{16}\cdot 4\text{H}_2\text{O}$ ), this phase also shows potentially interesting ion exchange properties.

### Thermal conversion of Sr-loaded IONSIV

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#### Abstract

The separation of radioactive Cs and Sr from waste streams has been an important process for the last 40/50 years. Many exchangers have been used or 'spent' and now await final conditioning and storage. In this study we focus on the consolidation of spent Sr-IONSIV R9120-B (formally IE-911), a commercial exchanger developed and engineered by Honeywell UOP, via hot isostatic pressing (HIPing).



# Metal substitution and Cs/Sr Ion Exchange in Sn-silicate Umbites and Thermal Conversion by Hot Isostatic Pressing

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## Abstract

Sn-silicate umbite ( $K_2SnSi_3O_9 \cdot H_2O$ ) crystallises in an orthorhombic system with the space group P212121 ( $a = 10.101$ ,  $b = 13.136$ ,  $c = 7.157$  Å) and is reported to ion exchange both Cs and Sr. Various metal doped microporous stannosilicates with an umbite structure of composition  $K_{2-x}Sn_{1-x}M_xSi_3O_9 \cdot H_2O$  (where  $x = 0.125 - 0.25$ , and  $M = Nb^{5+}$ ,  $Sb^{5+}$ ,  $Y^{3+}$  and  $Sc^{3+}$ ) have been prepared successfully via hydrothermal synthesis. It was found that the Cs and Sr uptake can be significantly improved by modifying the framework. Current work has focused on the structural studies on the Rietveld analyses of the synchrotron and neutron data. The systems with pentavalent metal substitution, e.g.  $Nb^{5+}$  and  $Sb^{5+}$  doping, structural analyses are consistent with no obvious change in the unit cell parameters or  $K^+$  bonding prior to the exchange, it is proposed that the improved property is due to the creation of cation defect sites within the pores of the material that facilitates greater cation mobility and leads to exchange. After ion exchange, a preliminary study of HIPing umbite materials is also presented aiming to obtain a dense wasteform.

# Assessment of empirical potentials for simulations of glass/crystal interfaces

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## Abstract

Empirical potentials have been tested in order to assess their usability in the study of glass/crystal interfaces of the type commonly found in nuclear glass wasteforms. Compositions considered in this work are  $(Na_2O)_x(SiO_2)_{(1-x)}$  silicate glasses and  $TiO_2$  crystals with the rutile structure (P42/mnm). Properties of both crystal and glass have been calculated to verify that the potential describes a realistic material. Finally, preliminary results of glass/crystal interface are shown.

# Temperature dependence of Cs volatilisation from Fukushima adsorbents

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## Abstract

For the remediation of the Fukushima site the volatilization behaviour of Cs from adsorbents is possible concern. Reliable data concerning Cs volatilization behaviour is required. For this work a novel experimental rig was set up and an effective and safe operating procedure was devised. A commercial zeolitic adsorbent was loaded in 1000 ppm Cs solutions to an effective wasteloading of 7-8 wt.% of Cs, and tested in temperature ranges from 600°C to 1000°C. The recovered Cs was analysed using Atomic Absorption Spectroscopy to determine the amount of volatilized Cs. It showed increasing volatilization at higher temperatures, with

behaviour following similar reported values in literature. The zeolitic adsorbent showed very little volatilisation with a relative volatilization of 0.097% at 1000°C.

## **Nanoparticle Technology in Waste Disposal and Remediation Systems**

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Imperial College, UK

The rapid development of the nuclear industry and the associated production of toxic waste has created a large demand for the design of new novel materials aimed at the removal of soluble metals and radionuclides for safe and efficient waste disposal.

The recent years' development of magnetic nanoparticles for separation technologies in liquid systems is widespread and already in use in medical testing. The challenge is to translate this methodology, via surface functionalization to target the species of interest, and to use superparamagnetic behaviour for waste form speciation and separation. The potential for magnetic-sorbent structures to be used in waste form separation or removal of RNs from liquid streams has been investigated. Particle development is being carried out and characterization studies performed using TEM-EDX, STEM-EDX, ATR-FTIR, XRD, DLS and  $\zeta$ -potential. Sorption tests with U and competing cations, such as Sr(II), Mg(II), Ca(II) and K have been carried out at pH 7 and pH 11 to evaluate the sample specificity. Preliminary desorption tests are currently being undertaken.

## **Developing the use of an Acoustic Backscatter system (ABS) to measure the concentration of nuclear simulants under varying degrees of flocculation**

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### **Abstract**

It is proposed here that further research efforts should be made in order to develop the use of acoustic backscatter to measure concentration in flocculated systems. The use of acoustics is generally widespread in marine, food and water treatment industries and has great potential in applications where access to process fluid presents a chemical or radiological hazard. Despite the fact that there have been many developments made recently by various authors to better understand how acoustic models can be developed to further to broaden their applications to measuring particle size and concentration in flocculated or otherwise cohesive systems there is not a robust model relating flocculated particle structure to its acoustic backscatter properties.

## **Gas release from weakly consolidated nuclear legacy waste**

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<sup>3</sup> Faculty of Engineering, Architecture and Information Technology, University of Queensland, Brisbane, Qld 4072, Australia

### **Abstract**

Retention of hydrogen bubbles within consolidated soft sediments represents an important safety consideration for the management of legacy nuclear wastes due to the potential for acute gas release. X-ray

computed tomography analysis of oxygen bubbles within low-intermediate strength (7-234 Pa yield stress) magnesium hydroxide soft sediments revealed that the largest mature bubbles exhibit surprisingly long residence times in the bed, which implies that continuous gas release is governed by a novel mechanism involving sub-millimetre bubbles rather than by the most buoyant population. A new x-ray tomography configuration at reduced scale and increased resolution has enabled the visualisation of a vast population of pore to millimetre scale bubble features which account for over 90 % of the bulk voidage. Three-dimensional image analysis has shown that these sub-millimetre bubbles grow with increased aspect ratios within more consolidated sediments, while these sub-millimetre bubbles are able to form extensive three dimensional networks, even within weakly consolidated sediments, thus enabling transport of retained gas over length-scales of many centimetres. The propagation of permeable bubble networks by capillary invasion is greatly influenced by the microstructure of the bed; a methodology is outlined for characterising pore size distributions of consolidated nuclear legacy waste test materials using a combination of nuclear magnetic resonance and mercury intrusion porosimetry, revealing pore throat radii in the range of 0.08-0.2  $\mu\text{m}$  within a 42.3 % w/w sample of corroded magnesium sludge.

## **Simulation of dense suspension in pipe flows with homogeneous stationary sediment bed**

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### **Abstract**

Dense suspensions in turbulent pipe flows with stationary variable flat-bed height are studied using large eddy simulation and discrete particle simulation, and compared to a reference full pipe flow. The flat boundary induced to some degree secondary flows and modifications to the fluid turbulence statistics and particle dynamics in the near-wall region. This work is motivated to simulate various degrees of bed formation during nuclear waste mobilisation and transport. Particle-particle interactions (collision and agglomeration) and particle-turbulence interactions (breakup of agglomerates) which can occur during some stages of the decommissioning process (retrieval and transport) of UK legacy nuclear waste, resulting in unwanted effects such as the formation of deposits and plugging of transfer and process pipes are considered in terms of this complex system.

## **In-line rheometry and flow characterisation of dense slurries in pipe flow using acoustic methods**

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### **Abstract**

The UK's civil nuclear waste inventory is large, diverse and requires processing and disposal but characterisation of nuclear waste slurries during storage, resuspension and hydraulic transport is made difficult by poor accessibility, radioactivity and chemical hazards. The aim of this study is to develop a suite of safe, accurate, versatile and cost-effective acoustic methods for rapid monitoring and characterisation of high-concentration nuclear waste-analogue slurries, based on existing expertise at the University of Leeds. Results from several acoustic methods are described: in-line pipe rheometry, bed depth and critical deposition velocity determination. A new flow loop laboratory has been commissioned in which these methods were combined with pumped sampling to investigate horizontal and vertical flow of suspensions of relevance to nuclear waste decommissioning and disposal.

# Quartz Crystal Microbalance as a Tool to Measure Complex Suspension Rheology

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## Abstract

The UK nuclear industry is currently entering a phase of post operational clean out, where the remediation of Sellafield, the largest nuclear site in the UK, is anticipated to cost £ 53 bn over the next 100 years. Substantial cost is associated with the clean-up and safe transfer of complex radioactive legacy waste sludge encountered in ponds, silos and highly active storage tanks across the site. To develop suitable design strategies for the mobilization and transfer of the sludge, its rheology should be accurately determined. This work demonstrates the use of a quartz crystal microbalance (QCM) to measure sludge rheology and compare the results to its shear yield stress. The device is simple to operate with no mechanical parts near the test material, is small and portable which allows for deployment into limited access areas and provides the user with in-situ rheological information on the test sample which eliminates the need for operator sampling. The measurement principle of the QCM relates to the changes in resonance frequency and motional resistance of a piezo-electric gold-coated quartz sensor as it is submerged into the desired test material. The air-to-sample frequency and resistance shifts are shown to correlate well with the shear yield stress of a suspension as measured by conventional vane viscometry.

# Application and theory of shear enhanced ultra-filtration with calcite suspension

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## Abstract

By applying shear to the surface of a filter during micro- and ultra-filtration the flux across the filter increases. Typical reports are of 4x increases, but reports exist of up to 17.1x (Jaffrin et al. 2004). Oscillation has been demonstrated to have superior shear generating conditions over rotational systems (Zamani et al. 2015) due to the inertia of the fluid, and simplicity of seals. This work investigates the core principles at work. A calcite suspension was tested under enhanced shear conditions to determine the fundamentals of this effect. A standard ceramic filter and a slotted nickel filter were tested. The filters were oscillated both axially and azimuthally (up and down, and around the axis). The cake build-up under various applications of shear was investigated, and compared to the particle retention, and any change in the resistance of the filter in question. This work investigates a predictive model of flux, dependent on shear and the suspension properties. This work shows that the filter cake provides both the particle retention and the largest resistance to filtration. Filter cake at pseudo-steady state is dependent on the magnitude shear stress, not application of shear (azimuthal/axial), filter used or concentration of source suspension. The thickness of cake, permeability, applicability to currently used theory, initial stages, and specific shear stress have been investigated intensively and backed up with computational fluid dynamics. The overall aim of this work is to build a method of determining an approach to determining pseudo steady state flux (and appropriate particle retention) knowing the shear stress and particle size distribution of suspension.

## Feasibility of Ultrasonic Spectroscopy for Use In-Situ Sludge Characterisation

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### Abstract

The aim of this study is to determine the feasibility of ultrasonic spectroscopy as a technique for in-situ measurements of particle size distribution of sludge. Current ex-situ techniques such as the laser diffraction method require a periodic collection of pond sludge samples and analysis in laboratories. In contrast, an in-situ solution is expected to improve data gathering efficiency, confidence in results and safety of sludge characterisation campaigns. Despite its suitability for high concentration sludge, the deployment of ultrasonic spectroscopy for analysing sludge in-situ may not be feasible. This is because of its dependence on a number of thermo-mechanical properties, a non-readily available data, particularly for legacy nuclear waste. Recent researches suggest this limitation could be by-passed if an analytical approach to data interpretation can be adopted, rather than the theoretically established mathematical approach. In furtherance, this paper suggests the use of existing mathematical theories to develop a series of training and testing data which may be used to develop an analytical model for data interpretation. Statistical analyses on arrays of related thermo-mechanical properties, resulting ultrasonic data and predicted particle size data will be employed. It is expected that the identification of a recurring pattern in the relationship between these data will be an addition to knowledge.

## Determination of Hydrogen Evolution from Legacy Fuel Storage Pond Sludges through Molecular Modelling

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### Abstract

The First Generation Magnox Storage Pond (FGMSP) represents one of the highest priority targets for risk reduction at the Sellafield Site. This legacy pond has accumulated a deep layer of sludge over many years, formed primarily from corroding Magnox alloy, but also from windblown debris and decaying organic matter. This sludge is of complex and uncertain composition. Additionally it contains dissolved fission products where cladding has failed and split, and even fragments of spent fuel where cladding has corroded entirely. The production of methane and hydrogen gas complicates the future handling and disposal of this sludge. The focus of the modelling project is the radiolytic route to hydrogen evolution through the study of a water-brucite-hydrocarbon system. By studying a simple model system we will develop an understanding of the underlying chemical reactions. Using density functional theory, we simulate radiation damage through the addition of excess electrons and holes. In this work, we show the effect these electronic defects have on bulk phase brucite mineral, which represents the most basic approximation to real Magnox sludge.

## **Thermal treatment of plutonium contaminated materials (PCM) waste**

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### **Abstract**

Key drivers for the application of thermal treatment processes include the reduced volume, improved passive safety, and superior long term stability, of the vitrified wasteform products. These advantages have led to a renewed interest in thermally treating various UK ILW streams. To support the increased investment in thermal treatment technologies a fundamental understanding of the processes and the impact of waste inventory needs to be established. The research described in this report aims to provide the evidence necessary to support a major investment in thermal treatment of plutonium contaminated materials. The report describes the use of laboratory scale waste simulants to develop an understanding of the waste and matrix interactions during thermal treatment of PCM waste. The report includes the thermal treatment process, characterisation and long term performance of the vitrified product.

## **Theme 4 – Structural Integrity**

### **Re-Use and Volume Reduction of Scabbled Contaminated Concrete from Nuclear Decommissioning**

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### **Abstract**

Around 50% of the UK's classified nuclear waste is building waste including concrete, cement and rubble. While research has gone into ways to minimise the volume of this waste, little has examined recycling of the materials. This research aims to investigate methods to reduce the burden on present and future storage and disposal facilities through re-use and volume reduction of conditioned contaminated concrete. Characterisation of ground, aged concrete has shown the possible presence of additional reactivity, while an investigation into the re-use of scabbled concrete within encapsulation grouts is ongoing. Laser Induced Breakdown Spectroscopy has been used to attempt to identify contamination products within a cement paste, with detection of a number of simulant radionuclides at inclusion rates as low as 0.1wt%.

### **Investigating the effects of ionizing radiation on Calcium Silicate Hydrates using First-Principles Calculations**

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### **Abstract**

The long-term storage of nuclear waste remains a significant challenge for the nuclear industry. One of the most vital materials employed in this storage process is cement – a complex, poorly understood calcium silicate hydrate mineral. Ab initio simulation of cement under the effects of ionizing radiation suggests that

excess electrons can solvate in the water-rich cavities and form radicals that may result in the production of H<sub>2</sub> gas. Electron holes also tend to localise at electron rich oxygens found in the cement structure and may further contribute to gas formation. This gas is highly dangerous as it is flammable and may cause pressurisation that could result in dangerous radioactive materials being released into the environment. To safely and effectively store waste, a deep understanding of cement and the interactions between it and the radioactive materials it may encounter is essential.

## **Production of biogenic phosphate minerals and applications for the remediation of radionuclides**

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### **Abstract**

The microbial production of phosphate minerals has gained attention as a promising mechanism of in situ groundwater remediation. To date, most research has focused on the immobilisation of contaminants by the induced precipitation of insoluble contaminant-phosphate minerals (e.g. uranyl phosphates); however, it is also possible to use this technique to manufacture materials such as calcium phosphates that can incorporate and immobilise a range of contaminants within their structure. These calcium phosphates can be utilised for the long-term, passive remediation of groundwater flow. This project investigates the use of fungi to induce the formation of calcium phosphate minerals via the enzymatic hydrolysis of an organic phosphate substrate (phytate). Attention is given to the factors influencing phytate degradation, biomineral formation, composition and characteristics, and this knowledge used to optimise biomineral production.

## **Use of colloidal silica grout for ground barriers in decommissioning: injection experiments and numerical simulations**

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### **Abstract**

Over the last three decades, colloidal silica has been investigated and more recently adopted as a low viscosity grouting technology (e.g. for grouting rock fractures within geological disposal facilities nuclear waste). The potential of colloidal silica as a favourable grouting material exists due to: its initial low viscosity; its low hydraulic conductivity after gelling (of the order of 10<sup>-7</sup> cm/s); the very low injection pressures required; its controllable set/gel times (from minutes to several days); the fact it is environmentally inert; its small particle size (less than hundreds of nanometres) and its cost-effectiveness. Colloidal silica can be destabilised by the addition of a salt accelerator compound and a change to pH, resulting in a rapid increase in viscosity (i.e. gelation) and formation of a rigid solid gel. This behaviour allows for low injection pressures to be used during the grouting process due to the initial low viscosity; with the resulting gel forming the contaminant ground barrier. This study aims to investigate the use of colloidal silica based grouts for formation of ground barriers at the Sellafield site. Potential applications include pre-treatment of the ground beneath, and surrounding, legacy structures prior to the retrieval of hazardous wastes and the formation of horizontal and vertical barriers surrounding unlined waste disposal trenches. In this summary some preliminary results on lab-scale injection experiments and finite element simulation are compared.

## **Smart cements for chloride / moisture sensing in nuclear concrete assets**

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### **Abstract**

Chlorides and moisture are two leading causes of reinforced concrete degradation. The issue is particularly important in a nuclear context, where concrete assets are usually coastal and may underpin safety-critical structures and radiation barriers. The goal of this research work is to develop an affordable, non-destructive, and combined monitoring and maintenance technology to support concrete integrity during operation and decommissioning in the presence of chlorides, water and radiation. We are developing novel “smart cements” coupled with an electrical system for use as a) chloride / moisture sensors and b) surface repairs for concretes at Sellafield. The project’s objectives are to: i) develop field-applicable sensors, ii) assess their sensing and repair performance under beta and gamma radiation, moisture and chloride contamination and iii) develop their associated EIS (Electrical Impedance Spectroscopy) interrogation and data processing systems. In this paper we present the first results of making geopolymers binders, putting them on concrete samples and curing them, for the particular application of monitoring.

## **The effects of colloidal silica based grouts on Sr and Cs speciation – present and future research**

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<sup>2</sup> Australian Nuclear Science and Technology Organisation, Kirrawee DC, New South Wales 2232, Australia

### **Abstract**

This study aims to investigate the use of colloidal silica based grouts for formation of ground barriers at the Sellafield site (Cumbria, UK) and for in-situ encapsulation of low-level wastes at the Little Forest Legacy Site (New South Wales, Australia). Here we summarise results on the interactions of radionuclides (Sr and Cs) with soil and waste materials. Finally, we summarize the pathways to obtain detailed information on the effects of colloidal silica based grouts on these interactions and the behaviour of colloidal silica based grouts during their injection.



