

DISTINCTIVE Work Package 3: Legacy Ponds and Silos Wastes

Bill Lee and Joe Hriljac,
Imperial College London and
University of Birmingham.

Sellafield Legacy Ponds and Silos: High Hazard Programmes.



Legacy Ponds & Silos



Pile Fuel Storage Pond



First Generation Magnox Fuel Pond



Magnox Swarf Storage Silos



Pile Fuel Cladding Silo



HAL (Highly-Active Liquor) Workstream

- **22% of all site programmes**
- **35% of total site costs during next 4 years**
- **77% of major project costs during next 4 years**
- **>90% of nuclear hazard potential on Sellafield site**

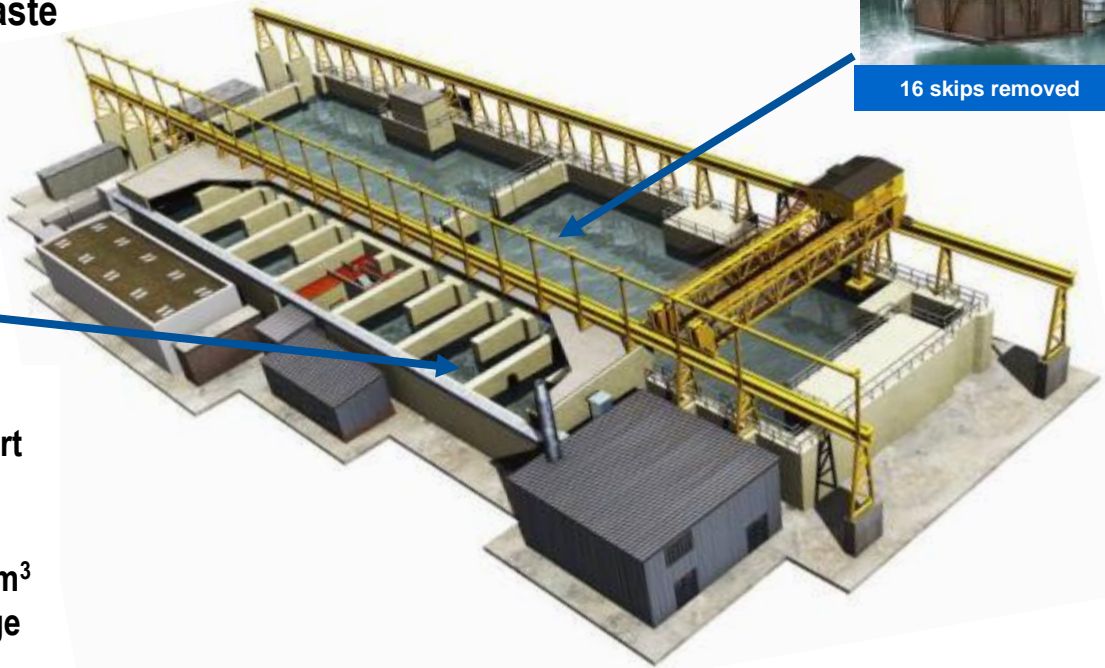
Pile Fuel Storage Pond

Legacy

- Constructed 1948 – 1952 to store, cool and prepare Windscale Pile fuel for reprocessing
- Waste consists of fuel, sludge and miscellaneous Intermediate and Low Level Waste



16 skips removed



- Sludge retrievals to an in-pond corral
- Local Sludge Treatment Plant* (LSTP) for short term storage of sludge
- Local Sludge Treatment Plant Process & Export* (LSTP P&E) to package sludge into 3m³ boxes and export for long term interim storage
- Oxide fuel to Oxide Fuels Storage Ponds for reprocessing
- Metal fuel to Fuel Handling Plant (FHP) for interim storage
- Remaining solid ILW inventory to pond solids conditioning facility, and packaged into 3m³ boxes for long term interim storage

- Bulk desludging to be completed during the 2015/16 FY
- Completion of the Local Sludge Treatment Plant Export Project planned for 2017/18 FY



DISTINCTIVE

NDA LP&S Strategy Objectives

- **Acceleration of High Hazard/High Risk Reduction**
- **Restore and maintain the basic condition of the assets and facilities.**
- **Reduce or mitigate the impact of the risk of a loss of containment of Nuclear Materials.**
- **Prepare the facilities for retrieval operations**
- **Retrieve the waste (hazards)**
- **Immobilise the waste (hazards), e.g. research into novel thermal methods.**

WP3: Aim and Objectives

- **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 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.

WP3: Resources

- **EPSRC funding for 12 years PDRA
(4 years each at Imperial, Birmingham and Leeds)**
- **12 DISTINCTIVE PhD students at:**
 - Imperial (Uni)
 - Loughborough × 2 (SL, Uni)
 - Leeds (SL)
 - UCL (NDA)
 - Bristol (SL)
 - Queen's University Belfast × 2 (NDA/QUB)
- **6 Additional PhD students at: Sheffield × 2 (NDA/Sheffield),
Imperial × 2, Birmingham, Leeds (CDT)**
- **First Theme Meeting held on Thursday 6th November in
Birmingham, ca. 35 attendees**

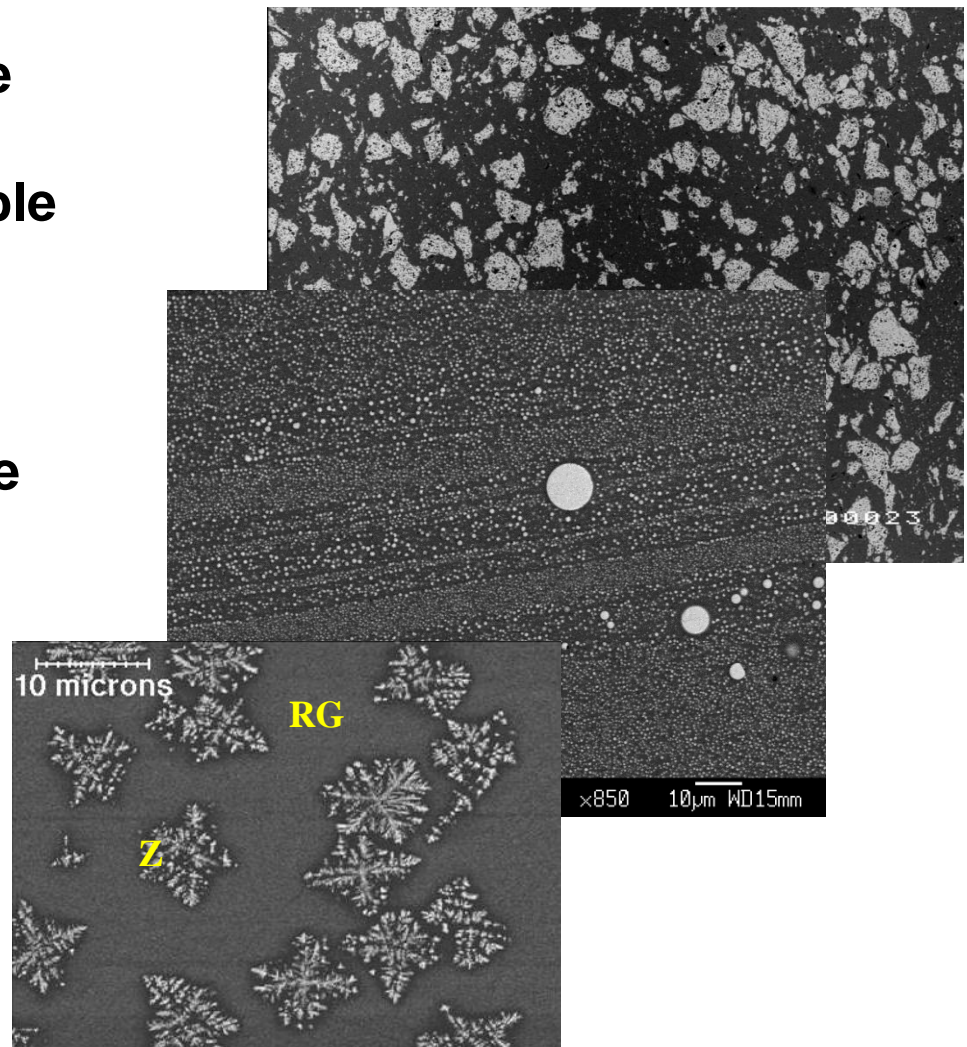
WP 3.3.1 Wasteform Durability

Lee & Grimes (Imperial), Hyatt (Sheffield)

- PDRA and 4 PhDs.
- Durability of Heterogeneous ILW Glass/Ceramic Wasteforms from Complex Wastestreams.(PDRA Lee/Grimes, James SL).
- Thermal Treatment Processes (2 additional PhDs Sheffield (NDA iCASE, NDA bursary, Hyatt, Maddrell NNL)
- Charlie Hutchison and Dimitri Pletser (2 additional PhDs Imperial)

Novel Wasteforms: Glass Composite Materials (GCMs) from Thermal Technologies

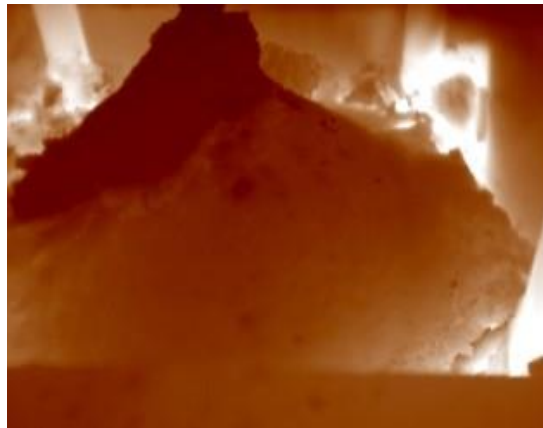
- Realisation over last decade that mixed crystal-glass wasteforms can be as durable as pure glass.
- E.g. crystalline waste encapsulated in melt which solidifies to glass (e.g. Joule Heater In-Can Vitrification).
- Applicable for some LP&S wastes.



WE Lee, MI Ojovan, MC Stennett and NC Hyatt, "Immobilisation of Radioactive Wastes in Glasses, Glass Composite Materials and Ceramics," Adv. Applied Ceramics 105 [1] 3-12 (2006).

Proof of Concept Trials using Surrogates

- Demonstrated potential of thermal treatment options to treat several LP&S wastes and they
 - can handle Sellafield LP&S wastes
 - are deployable at Sellafield
 - produce a durable product
 - offer cost benefits
- E.g. Joule Heater In-Can Vitrification – Mixed solids & sludge waste



Key Issues of Thermal Processes

- **Convert reactive material (e.g. metals, sludges & organics) to more stable forms. But the following need addressing:**
 - **Variable nature of wastes make control of process and product difficult.**
 - **Difficult to characterise heterogeneous waste and product.**
 - **Durability testing of product.**
- **PDRA will model aqueous corrosion of heterogeneous wasteforms from novel thermal processes and link with experimental programmes on durability at Sheffield and Imperial.**

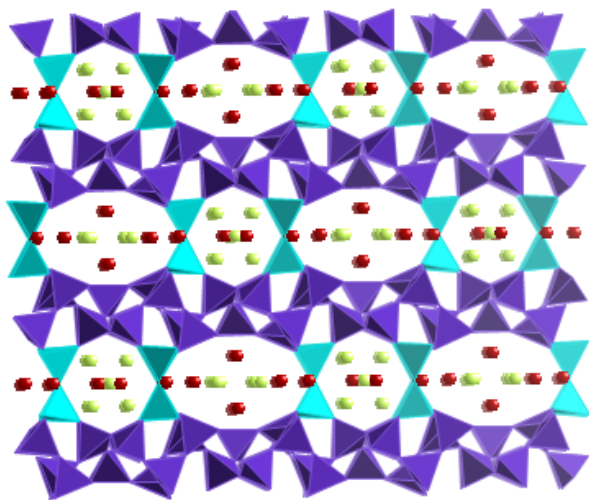
WP 3.3.2 Effluent Treatment and Analysis

Hriljac & Read (Birmingham), Evans, Platt , Dragosavac & Holdich (Loughborough), Ryan & Vandeperre (Imperial)

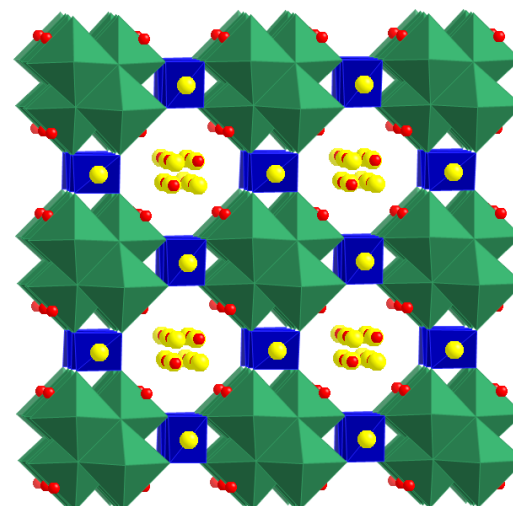
- **PDRA and 4 PhDs.**
- **Novel Ion Exchange Materials (PDRA + additional PhD, Birmingham, Hriljac/Read, Kellet SL).**
- **Magnetic Nanoparticles for Waste Separation or Sequestration (PhD Imperial, Ryan/Vandeperre, O'Brien NNL; PhD Loughborough, Evans/Platt, Kellet SL)**
- **Enhanced Shear Micro- and Ultra-filtration Without Recycle Pumping (PhD Loughborough, Holdich/Dragosavac, Kellet SL)**

Ion Exchange Materials

- Effluent clean-up for removal of soluble radionuclides such as Cs-137 and Sr-90 traditionally rely on an ion exchange process with a porous inorganic solid



zeolite clinoptilolite
 $(\text{Na}, \text{K}, \text{Ca})_3\text{Al}_3\text{Si}_{15}\text{O}_{36} \sim 10\text{H}_2\text{O}$



Acid form of CST in IONSIV
 $\text{Na}_{0.4}(\text{H}_3\text{O})(\text{Ti}_{1.4}\text{Nb}_{0.6})\text{O}_3\text{SiO}_4 \sim 2\text{H}_2\text{O}$

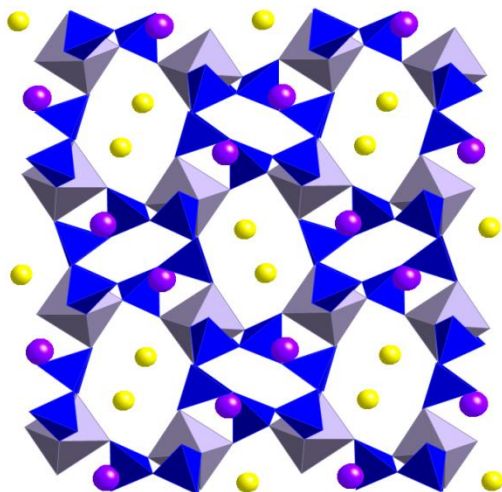
Ion Exchange Materials

Limitations of current systems include

- **Variability of source for natural materials such as clino**
- **Cost, e.g. IONSIV contains substantial amounts of Nb (in a 1995 Hanford clean-up of 187,000 m³ of liquid HLW ca. \$163M was allocated to purchase the IONSIV)**
- **Efficacy under acidic conditions**
- **Unknown efficacy in the presence of complexants and/or decontamination agents**
- **Potential for back exchange under storage conditions**
- **Conversion routes to good wasteforms for long term storage**

Novel Ion Exchange Materials

- Use atomistic modelling to pre-screen mineral phases to predict best candidates for ion exchange
- Make phases via hydrothermal and/or microwave routes
- Characterise and test for ion exchange properties
- Investigate thermal processing routes including HIPing to make dense wasteforms



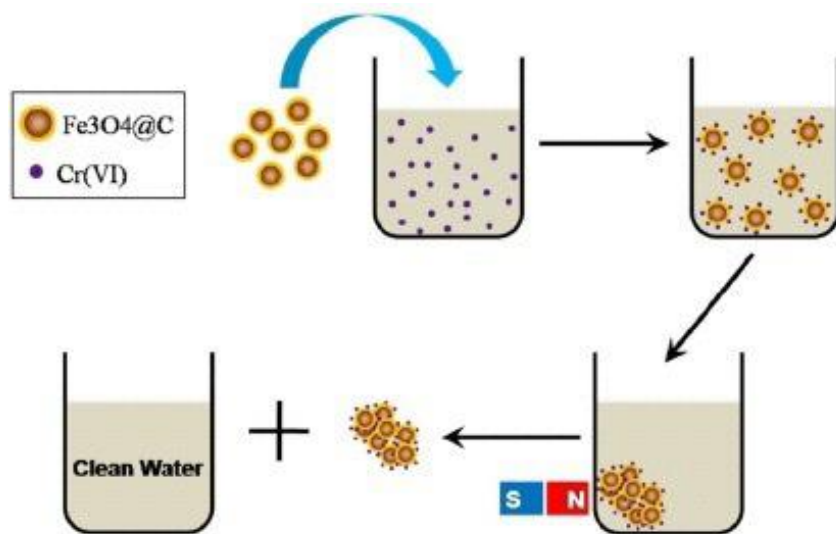
- $(\text{Na,K})_2\text{SnSi}_3\text{O}_9 \cdot \text{H}_2\text{O}$
- K_d for Sr nearly as good as IONSIV and SIXEP clino
- Thermally converts to a dense phase
- Work of current PhD student Sav Savva (NDA bursary)

Magnetic Nanoparticles for Waste Separation or Sequestration

Project 1

Developing Magnetic NP Technology for Sensing & Separation

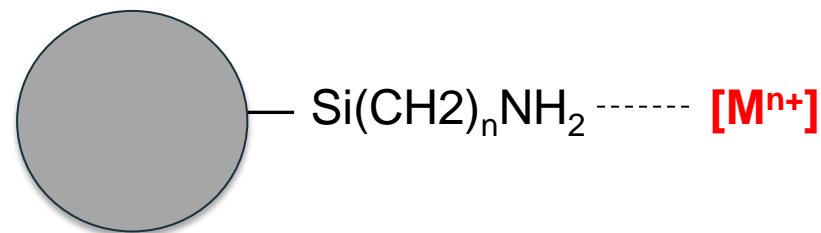
Mary Ryan & Luc Vandeperre



Project 2

Applications of Nanopore Technology for Ultra-sensitive Detection

Mark Platt & Nick Evans

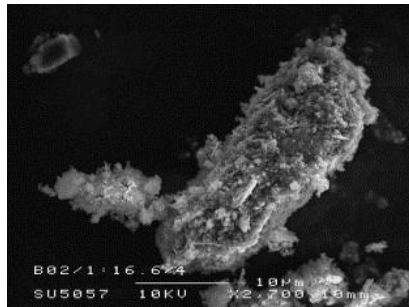


Use Resistive Pulse Sensing to detect metals bound to particles after they are magnetically separated from solution

Enhanced Shear Micro- and Ultra-Filtration Using Oscillatory Flow Conditions

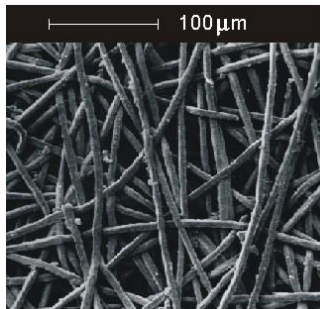
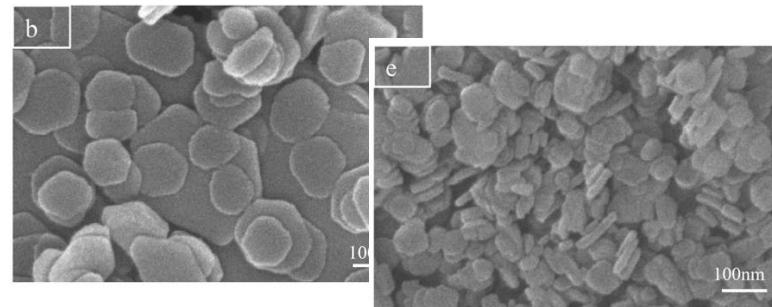
Radioactive ferric flocks

- Easily broken down
- use oscillating membrane

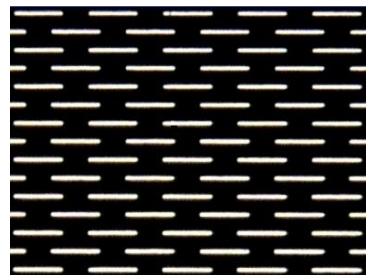


Magnesium hydroxide

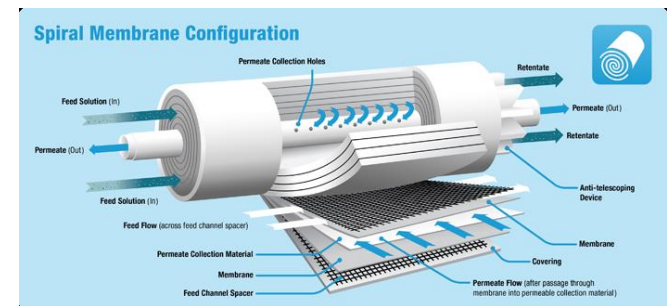
- Stronger particles
- Use pulsation of suspension



**Conventional
Microfilter, 3 micron**



**Metal Filter
2 micron available**



Spiral wound membrane

WP 3.3.3 Pond and Silo Sludges

Day & Scott (Bristol), Fairweather, Peakall, Hunter, Harbottle & Rice (Leeds), Currell, Kohanoff & Tribello (QUB), Kerridge (Lancaster).

- PDRA and 6 PhDs.
- Measurement and Modelling of Sludge Mobilisation and Transport (PDRA Leeds, Fairweather, Randall SL).
- Characterisation of flocculated waste suspensions with acoustic backscatter (PhD Leeds, Hunter/Harbottle/Peakall/Rice, Randall SL)
- Gas Hold-up in Sludges (additional PhD Leeds, Peakall, Randall SL)
- Computational Simulations of Storage Pond Sludge Disturbance (PhD Lancaster, Kerridge, Owens NNL)
- Development of Raman Spectroscopy Techniques for the Remote Analysis of Nuclear Wastes in Storage (PhD Bristol, Day/Scott, Rogerson SL)
- Experimental and Theoretical Studies of Irradiated Sludges (2 additional PhD's QUB, Currell/Kohanoff/Tribello, Barnes SL)

Measurement and Modelling of Sludge Mobilisation and Transport

Current understanding of behaviour of nuclear waste sludge during mobilisation and transport poor due to:

- **Complex nature of particle phase (size distribution, shape, density, etc.)**
- **Limited availability of useful data for these materials**

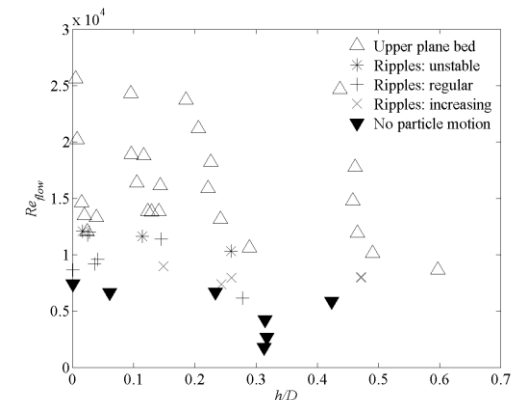
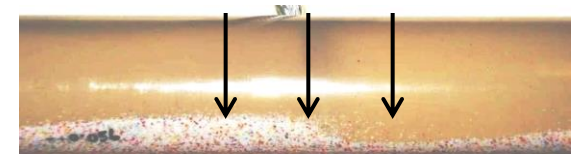
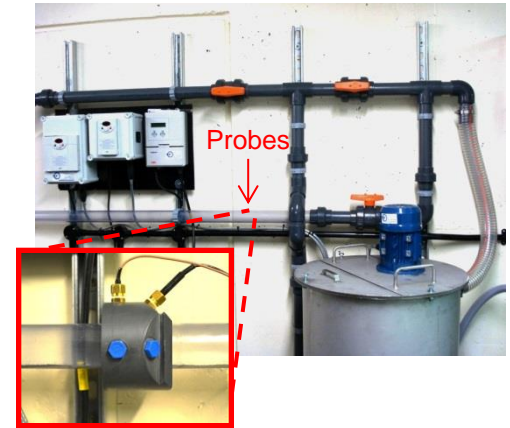
Further understanding of sludge materials, and development of quantitative predictive procedures, necessary for:

- **Design of more efficient and safer treatment processes**
- **Reductions in cost and operational timelines**
- **Improved understanding of operational problems**



Measurement and Modelling of Sludge Mobilisation and Transport

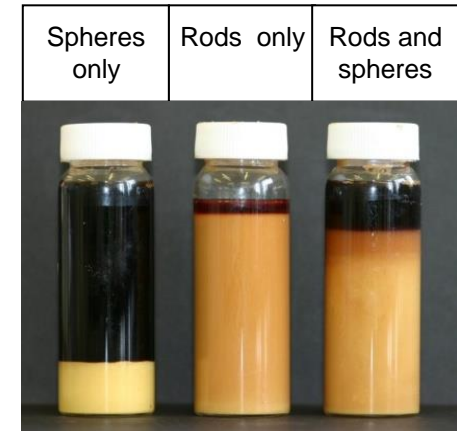
- Linked work to be undertaken by PDRA(s) on two projects
- First project will use realistic sludge simulants (recipes from NNL/Sellafield):
 - With range of particle shapes, sizes and densities
 - Examine behaviour of particle-laden flows, at various solids concentrations, in horizontal and vertical pipes
 - Characterise segregation behaviour of particle materials using novel ultrasonic approaches



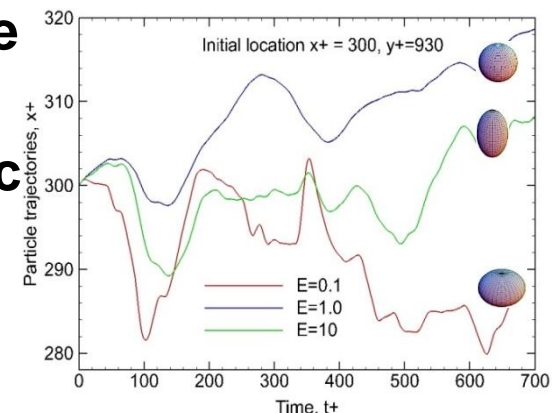
Bedforms in turbulent pipe flow

Measurement and Modelling of Sludge Mobilisation and Transport

- **Second project will use coupled large eddy simulation/Lagrangian particle tracking to simulate pipe flows and:**
 - **Influence turbulence and direction gravity on particle agglomeration, settling, and shear break-up**
 - **Range of particle shapes, sizes and densities**
- **Overall aim to provide validated method for complex flows in geometries relevant to waste processing operations**
- **Results of benefit as touchstone for pragmatic models used by industry**



Settling behaviour of different shaped particles



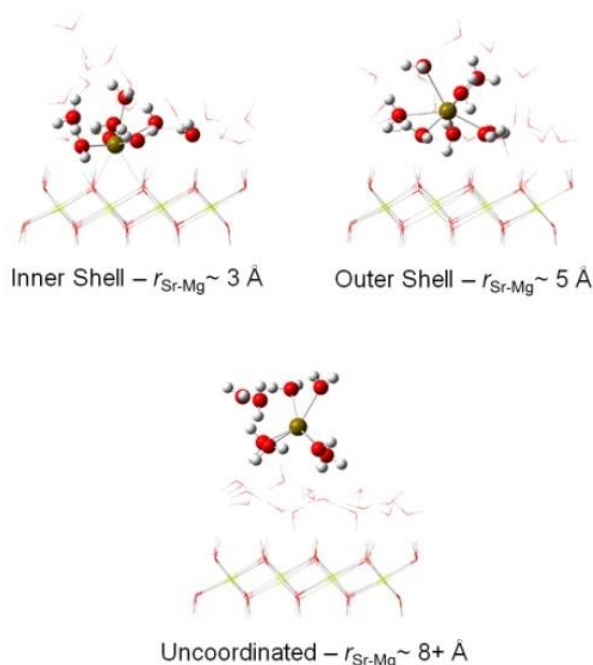
Trajectories of different shaped particles in flow

Characterisation of Flocculated Waste Suspensions with Acoustic Backscatter

- **Aim is to quantitatively understand the flocculation of fine magnox and the analysis of settling sludge systems with the acoustic backscatter array (ABA).**
- **First research strand is studying the flocculation of fine magnesium hydroxide particles with different commercial polymer agents.**
- **Second strand is to measure the fundamental relationships between acoustic scattering length, attenuation and velocity from these complex multicomponent species to aid in developing analytical methods for the ABA.**
- **Project will focus on linking theoretical scattering-attenuation relationships for use with flocculated waste suspensions.**

Computational Simulations of Storage Pond Sludge Disturbances

- Main aim is to better understand the interactions of Sr, Cs and U with water, hydroxide and the [0001] reactive surface of brucite
- Follow on work from Diamond consortium



- Embedded point charge brucite
- Solvation using 36 water molecules
- Three co-ordination environments:
 - Inner Shell
 - Outer Shell
 - Uncoordinated

Figure 2 The three different coordination regimes exhibited by Sr²⁺ hydrates.

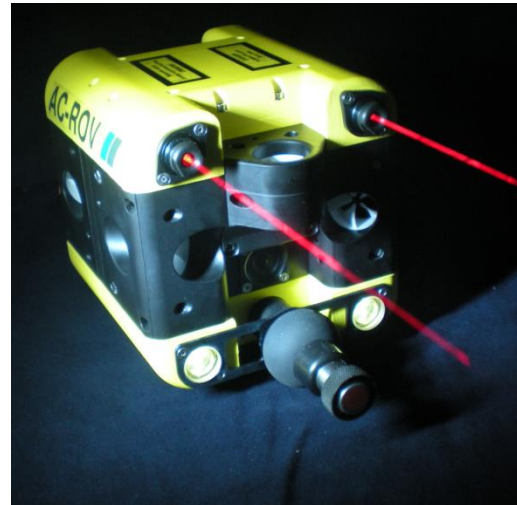
Raman Spectroscopy Techniques for the Remote Analysis of Nuclear Wastes in Storage

To develop improved techniques for remote monitoring of sludges and heterogeneous wastes

- **Development of laser and fluorescence imaging techniques for remote identification of key waste products in wet places.**
- **Miniaturisation and integration of prototype sensors with existing mobile platforms (ROVs, snakes).**

Underwater trials in Bristol and then hopefully ponds

- **Based on UoB excellence in sensor integration with mobile platforms, miniaturised Raman probes and fluorescence imaging units.**



Irradiation Studies of Sludges

- **PhD 1 – irradiation of sludge samples and assay of radiolytic products**
- **PhD 2 – atomistic simulations of the interactions between radiolytic products (particularly those found in high concentrations within the first microsecond of interaction with ionizing radiation) and clay - water interfaces.**
- **The outputs from the two sides of the project will be combined in a Monte Carlo framework to provide a predictive model of the radiochemistry of sludges.**