Understanding Wasteforms from Thermal Treatment Methods.

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Outline

• Controlled, uncontrolled/difficult wastes.
• UK Wastes: e.g. Sellafield Legacy Ponds & Silos (LP&S).
• Waste Immobilisation.
• Non-thermal Treatment of Waste.
• Thermal Treatment.
• Gaps and Opportunities.
Management of Controlled Wastes.

- From civil nuclear power and medical isotopes.
- HLW – reprocessing/vitrification or SNF.
- ILW – cementation or yellow boxes.
Wastes may be Difficult because they:

- Contain highly radiotoxic radionuclides emitting high-energy (α) radiation ($^{239}$Pu, $^{241}$Am, $^{237}$Np).
- Contain radionuclides with long half lives ($^{14}$C 5730y, $^{239}$Pu 24110y, $^{129}$I 15.7My, $^{99}$Tc 213000y).
- Contain highly mobile (water soluble or volatile) radionuclides (gases: $^{226}$Ra, $^3$H and $^{14}$CO$_2$, alkalis: $^{137}$Cs, $^{90}$Sr, halogens: $^{36}$Cl, $^{129}$I).
- Contain radionuclides easily assimilated with long biological half lives ($^{129}$I thyroid, $^{90}$Sr bones).
- Have escaped into the environment (Fukushima, Hanford, Chernobyl, Dounreay) as mobile species/particles.
- Are high volume (graphite).
- Are in uncharacterised sludges (LP&S).
Uncontrolled/Difficult Wastes:

Global need for techniques capable of immobilising ill-characterised, heterogeneous wastes.

- Hanford (USA), Chernobyl (Ukraine), Fukushima (Japan), Dounreay (Scotland), Sellafield LP&S (England) etc.

Sellafield Legacy Ponds and Silos: High Hazard Programmes.

- 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

Baseline Plan

• 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
Pile Fuel Storage Pond

• Clearing Sludge from Pond sections.
• Completing storage facility construction.
• Removing redundant skips.
What is Immobilisation?

- Important to make the waste into a stable and durable form.
- Convert liquids and gases into solids.
- **Immobilise** waste by chemically incorporating into the structure of suitable matrix (glass or ceramic) so it is captured and unable to escape.
- **Encapsulate** or physically surround waste in material which isolates it and retains radionuclides (e.g. cement).
Immobilisation in Glasses.

- Glass structure has short range order but no long range order.
- Silicate tetrahedra link up to form 3D glass network. Some ions such as Na modify the network but are not part of it.
- Active species can go into the network or other sites. E.g alkalis ($^{137}$Cs, $^{90}$Sr) into Na site.
- Open and random structure can accommodate most of the Periodic Table.
Immobilisation in Ceramics

- Crystalline structures can accommodate radionuclides in very specific locations.
- Tunnels between $\text{TiO}_6$ octahedra in hollandite accommodate $^{133}\text{Ba}$, $^{137}\text{Cs}$ and $^{90}\text{Sr}$. 
Crystals in glass used to be unacceptable but now realise presence of crystals may not compromise durability.

GCM’s now accepted as wasteforms e.g. French Cold Crucible Melter active line at La Hague for U/Mo wastes started early 2010.

Crystal-tolerant glasses (with higher waste loading) to be used for Hanford wastes in USA.

E.g. glass ceramics or crystalline waste encapsulated in melt which solidifies to glass (e.g. Joule Heater In-Can Vitrification).

Applicable for some LP&S wastes.
Non-thermal Waste Immobilising Technologies.

- Much interest in near room temperature ceramics for ILW immobilisation.
- Possible workshop on these in 2014?

Thermal Waste Immobilising Technologies

- Many discussed in detail later today.

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
Characterisation and Durability Testing of Simulant Legacy Pond and Silo Waste.

- Charlie Hutchison, PhD  
  Imperial started Oct 2012.

- Examining several Glass Composite Material (GCM) samples from LP&S type wastes simulant trials using Joule Heated In-can Melting and Plasma Vitrification.

- Characterisation of phases before and after MCC-1 tests
  - Current: Optical, XRD, SEM, EDS, ICP and chemical analysis
  - Planned: ToF-SIMS, and cross sectional TEM.
Asbestos based GCM JH-ICV Sample

- Asbestos, a magnesium silicate material used for its thermal insulation and mechanical properties
- Vitrified but partially crystallised bright phases which EDS indicates are Ca-rich crystals surrounding cristobalite so a GCM.
Future Work.

• Further examination of different leaching down cracks compared to surface, effect of *local* equilibrium.

• To apply tests done to asbestos sample to all other GCM’s supplied by Sellafield

• Examine modelling options to help interpretation of experimental results

• Examine effect of iron on durability; closer to repository conditions.
Communal Opportunity.

• Dimitri Pletser (PhD Imperial started Oct 2013)

• Characterising other GCM samples from JHIM and plasma melting trials.

• Could pursue similar study to Charlie Hutchison but may also branch out, an opportunity to link with other groups here today.
Decommissioning, Immobilisation and Storage Solutions for Nuclear Waste Inventories (DISTINCTIVE) consortium

- Simon Biggs (Leeds, PI), EPSRC-funded with National Nuclear Laboratory, Sellafield, NDA etc.
- 4 themes: Structural Integrity, AGR, Magnox and Exotic Spent Fuels, PuO$_2$ and Fuel Residues, and Legacy Ponds and Silo Wastes.
- Supports 10 PDRAs and up to 20 PhDs.
- PDRA at Imperial on Glass/Crystal-containing Wasteforms.
Research Associate in Glass/Crystal Containing Wasteforms

• To be advertised this week on Imperial Dept. Materials website, starts Feb 2014.

• Working with Profs. Bill Lee and Robin Grimes, Dr. Mike Rushton and PhD students Charlie Hutchison and Dimitri Pletser at Imperial plus colleagues at NNL, NDA and Sellafield.

• Linked modelling and experimental study of wasteform corrosion in aqueous store/repository environments.
Novel Thermal Immobilisation
Gaps and Opportunities.

- Technology issues – processes, mobile units (accidents), applicability to differing wastestreams. Manufacturers (Kurion, Tetronics).

- Wasteform issues – ill-characterised wastes, heterogeneous wasteforms, durability. Users (Sellafield, NDA).
Key Wasteform Issues from Thermal Processes.

• Have converted reactive material (e.g. metals, sludges & organics) to more stable forms.

• However, the following need addressing:
  – Variable nature of wastes make control of process and product difficult.
  – Difficult to characterise heterogeneous waste and product.
  – Protocol for durability testing of heterogeneous product.
  – Need to make safety case for disposal of such wastes

• Nonetheless, the reduction in hazard is enormous and we need to be pragmatic.
For more information: