Polymer Encapsulation of Nuclear Waste: Alternatives to Grout

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Introduction

- UKAEA Waste Immobilisation Laboratories
  - Harwell and Dounreay
- Provide wasteform development services
  - Initial small scale tests right up to full scale trials
  - Generally in support of Letter of Compliance submissions
  - Ongoing plant support
- Traditionally used cement but things are changing

Cement

- Ordinary Portland Cement (OPC) is the preferred encapulant for nuclear waste in the UK.
  - Generally mixed with additives such as Pulverised Fuel Ash (PFA) or Blast Furnace Slag (BFS)
- Advantages
  - Flexible: Formulations can be easily tailored to individual wastes if required
  - Compatible with most wastes
  - Provides a good physical barrier to radionuclide release
  - High pH provides a good chemical barrier to release
  - Can be expected to be stable for hundreds of years
  - Good radiation tolerance
  - Porosity allows for release of gases
  - Cheap and reasonably readily available

Cement

- So why not use cement for everything?
  - Porosity
    - Allows access for water and leaching of radionuclides
    - Release of undesirable gases, eg. radon
  - Some wastes are incompatible
    - Aluminum
    - Uranium
    - Magnox
    - Ion exchange resins
  - Possible future issues with consistency of supply

Alternatives

- Other possible encapsulants include
  - Other inorganic systems
    - Calcium aluminate cements
    - Magnesium phosphate cements
  - Inert binders
    - Clay – LoC obtained for use at Harwell
  - Low melting point metals
  - Organic polymers

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Polymers

- More options
  - Thermoplastic?
    - Polyethylene
    - Polystyrene
    - Bitumen
  - Or Thermosetting?
    - Epoxy based systems
    - Polyesters

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Historical & Current Use

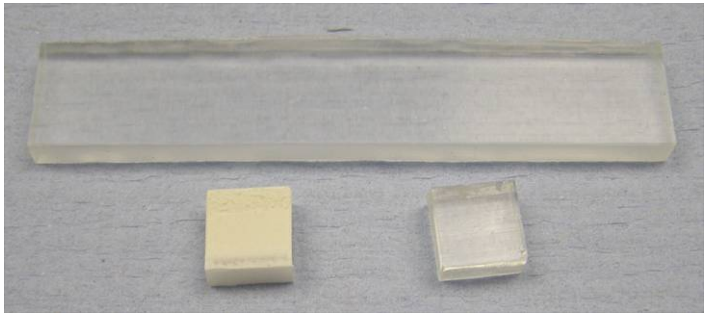
- UK R&D work in the ’80s
  - Cement generally chosen above polymers at the time.
  - Dissolution was favoured above the use of epoxies for treatment of Magnox FED.
- Only UK large scale plant to use polymer is Trawsfynydd
  - Encapsulation of ion exchange resins
  - Uses Vinyl Ester Styrene (VES)
- Harwell Radium Wastes
  - Small scale (up to 5 litre) encapsulation with VES
  - Then encapsulated in grout in 500l drums with other solid waste
  - Better encapsulation and reduced radon emissions
- Use elsewhere in the world
  - France, Germany, USA, Canada, Japan etc.
  - Using polyesters, epoxies, polyethylene etc.
  - Often for treatment of ion exchange resins

Current Investigations

- Ion exchange resins
  - Dounreay and Harwell
- Graphite dust
  - Achieves higher loadings than cement
- GLEEP Fuel
  - Aluminium clad natural uranium
- Oils in Imbiber Beads
  - Minimise leaching
- Wet wastes?
- Windscale Piles

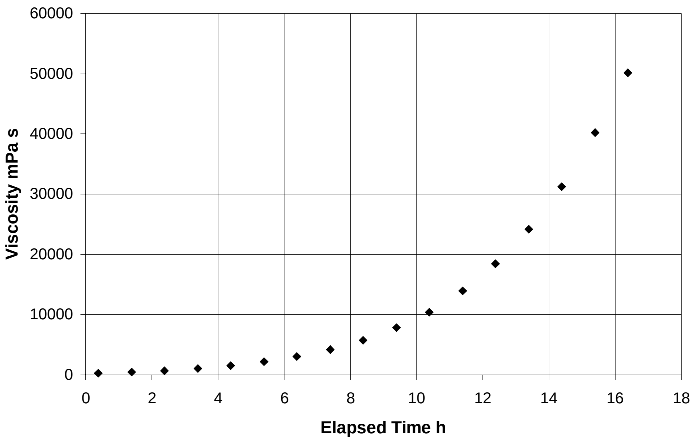
Windscale Piles

- Fuels and isotopes waste
  - Contains a significant quantity of metallic uranium making cement less favourable
- Various polymers investigated
  - Vinyl Ester Styrene (VES)
  - Advanced Polymer System (APS) epoxy
  - Alchemix epoxy
  - Huntsman epoxies



Properties of Polymer

- Compressive, tensile and flexural strength
  - Generally stronger than cement
  - Achieves maximum strength quickly
  - Both brittle and plastic failure possible
- Gelation and setting times
  - Largely depends on temperature
- Viscosity
  - Initial Newtonian behaviour
  - Exponential increase in viscosity as curing progresses
- Dimensional Changes
  - Polymers shrink during curing
- Leaching performance



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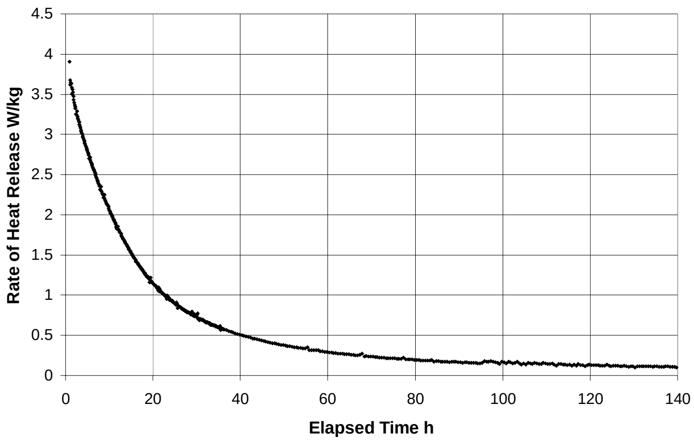
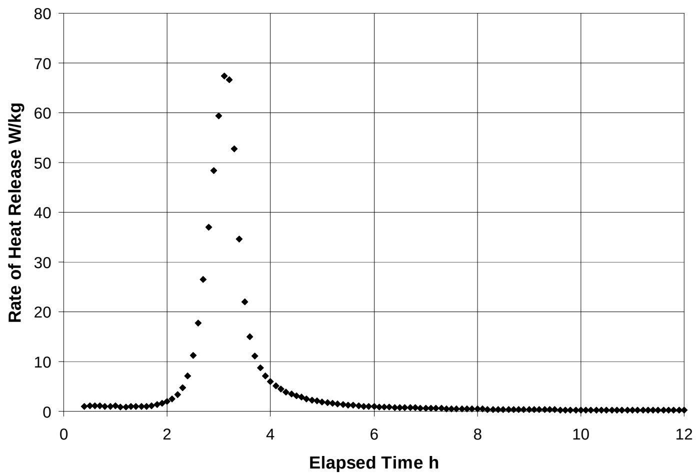
Heat Release

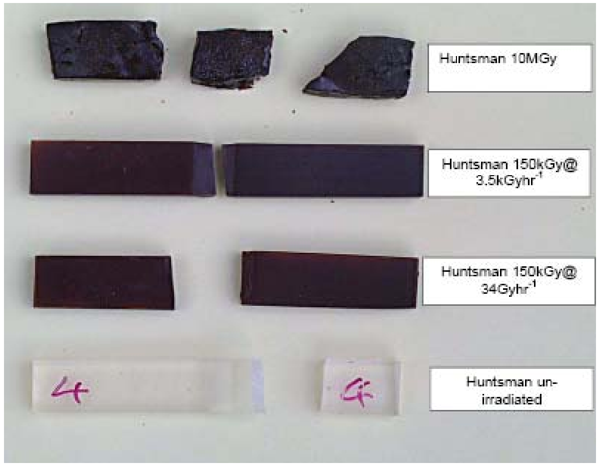
- Heat release / curing exotherm
  - Isothermal conduction calorimetry
  - Scaling trials from 500g to 25kg
  - Full scale (up to 300l) trials
- Lower heat capacity and thermal conductivity than cement
  - Cement heat capacity approximately twice that of polymer
  - Cement thermal conductivity ~5x that of polymer
- High temperatures achieved at larger scale (>200°C)
  - A possible issue with large scale polymer encapsulation
  - Cracking occurs due to internal temperature gradients and variation in the degree of cure
- Lead to investigation of “low temperature” formulations



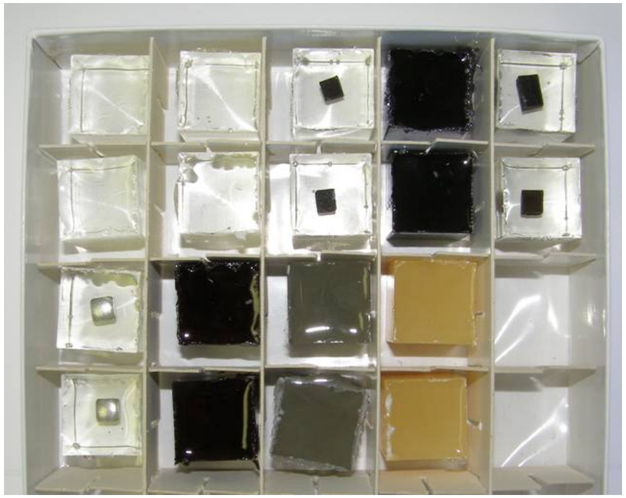


Radiation Stability

- Alpha irradiation and “tunnelling”
  - Two dose rates and a concentrated beam
- Gamma irradiation
  - 10 MGy at 3.5 kGy/h,
  - 150 kGy at 3.5 kGy/h and 34 kGy/h
- Tests performed
  - Three point bend and compressive strength
  - Infra-red spectroscopy
  - Gel fraction / solvent uptake
  - Leachate analysis
  - Gas generation
- Progressive changes in properties under irradiation
  - One polymer heavily degraded at high dose



Simulant Interactions



- Variety of simulants being investigated
  - Aluminium
  - Mild Steel
  - Graphite Powder
  - Isotope average and worst case mixtures
  - Depleted uranium
  - LiMg alloy
- Monitoring for 2 years
  - Currently just over 6 months in
- 5mm simulant pieces in 19mm polymer cubes
  - APS, Alchemix and low temperature Alchemix
  - Monitored for dimensional changes, compressive strength, and any visual observations
  - Sectioned and analysed by optical microscopy, SEM, DSC and TGA.
- Gas generation
  - 200ml polymer with equivalent simulant loading
  - Testing for oxygen, nitrogen, carbon dioxide, carbon monoxide, propane, hydrogen, methane, ethylene, ethane and acetylene.

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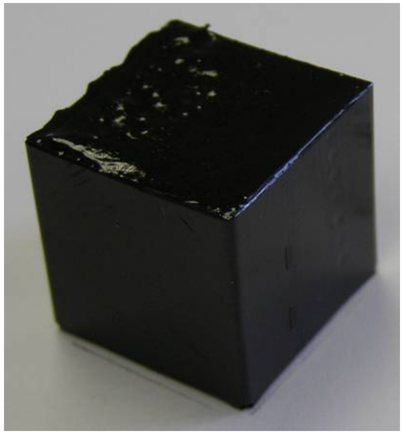
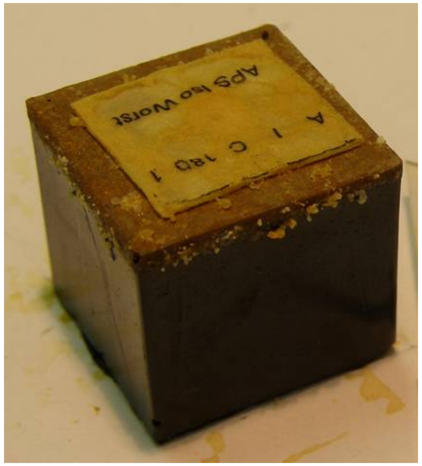
Results to Date

- To simulate conditions in a full scale drum, some polymers were cured at 180°C
  - Surface degradation
  - Production of CO_2, CO, CH_4 and C_2H_4
- Polymer has expanded slightly over time
  - Gains weight due to absorption of water from the atmosphere
- Isotope simulant sample show some damage over time
  - Reaction with humid storage environment to form metal hydroxides and carbonates
  - Efflorescence on base of cubes
  - Less CO_2 produced
  - Larger weight gains than other simulant cubes





Uranium Corrosion

- An important reason for using polymers
- Polymers do contain some water
  - Water content tests on components and set product (around 0.2%)
  - Some corrosion likely to occur, especially in absence of oxygen
- Hydrogen has been produced, though only in relatively small amounts
- 180°C cured samples exhibit a friable powder coating on the DU cube
  - Samples cured at 70°C are not exhibiting this
  - Corrosion appears to mostly occur in this initial high temperature phase.
  - Not on the same scale as with cement

Summary

- Polymers have many desirable features as encapsulants
  - High strength
  - Low permeability
  - Compatibility with ‘difficult’ wastes
  - Radiation tolerant
- Curing exotherm may be an issue for larger scale processes
  - However, full scale (up to 300 litre) trails performed by WPDP have been sucessful.
  - Low temperature formulations under investigation
  - Fillers?
- Some simulant effects have been observed
  - Isotope simulants
  - Corrosion of uranium at high temperature
- Overall positive at this stage
  - Work is ongoing

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Acknowledgments

- Thanks to
  - John Clifford, Sellafield Ltd
  - The staff of the Liquid Effluent Treatment Plant, Harwell

- Any questions?

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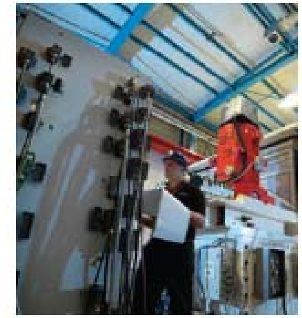


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