The REFINE Research Consortium

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4th NADM Sheffield- 9th Sep 2015

REFINE Consortium

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- The REFINE consortium
  - led by the University of Edinburgh
  - includes the Universities of Cambridge, Manchester, Nottingham and University College London, in partnership with UK National Nuclear Laboratory.
- a UK multidisciplinary programme
  - Chemistry, Engineering, Materials, Earth Sciences.....
- aimed at delivering essential pyrochemical
  - platform technologies
  - underpinning process development
  - training
- for safe, dependable and sustainable UK nuclear fuel reprocessing

REFINE programme

- REFINE is developing ……
  - specific sustainable spent fuel reprocessing technologies to produce a viable molten salts based, spent fuel treatment facility
    - minimizing waste
    - Legacy fuel reprocessing
    - for new Gen III+ and Gen IV reactor systems
    - delivering safe, reliable, economic and sustainable nuclear energy on the scale required in both the short and long term.
  - The programme is focussed on establishing the fundamental research, understanding and essential systems required.

REFINE Academics



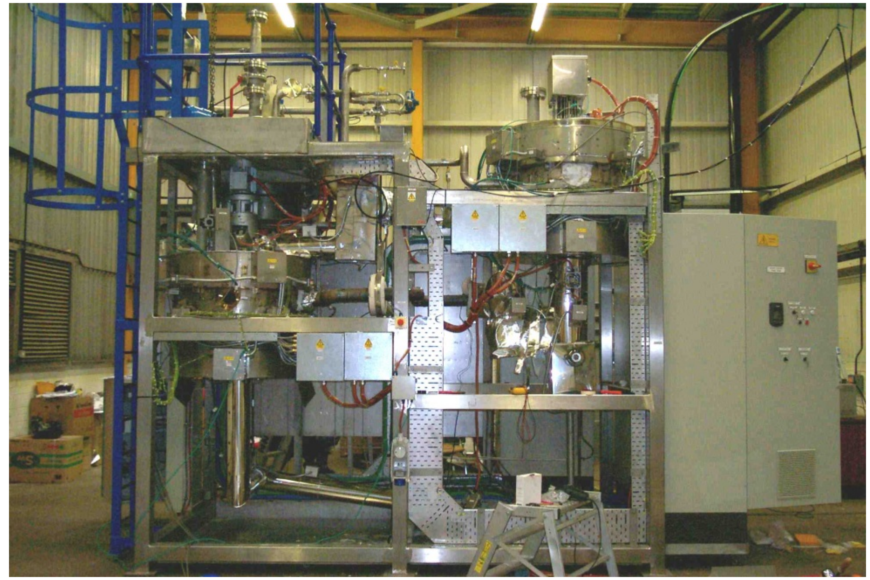


National Nuclear Laboratory

- Key Partner in REFINE
  - NNL contains a number of critical skills and facilities essential to support the nuclear industry in the UK
  - NNL has a key objective to help safeguard and develop nuclear expertise and multifunctional nuclear laboratory facilities
    - MSDR
    - Active experiments to benchmark surrogates

Molten Salts Dynamic Rig

- Designed, Tested and Commissioned by NNL Engineering Molten Salts Group
- Operating Temperature up to 500 °C
- 110 Kg Salt Inventory
- Argon Gas Atmosphere
- Range of Transfers Possible - Gas Lift, Fluidic, Mechanical Pump, Pressure, Gravity
- Pumping Rate > 4 m^3/hr
- Removable Sections for Testing In Line Components



MSDR held in a quiescent state for several years. Under REFINE, it has been re-commissioned and characterised.

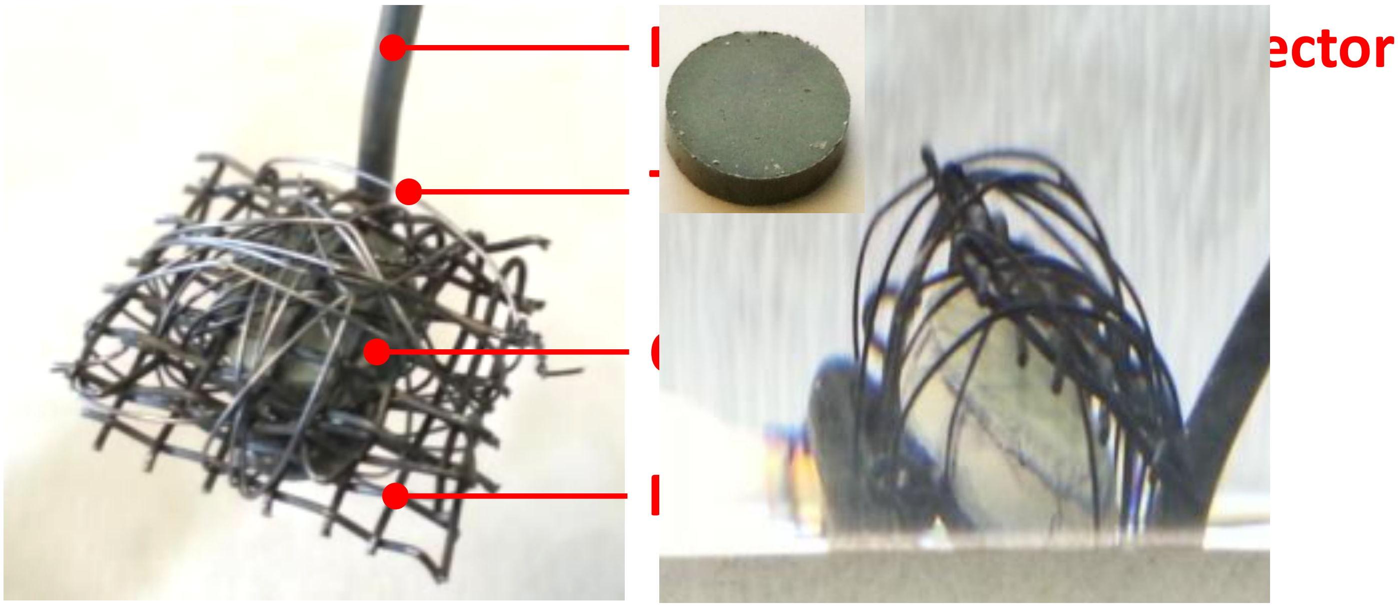
REFINE Technical Objectives

- In 4th year of 4.5-year programme:
- 3 key themes, workpackages, *objectives*
- Direct Electrochemical Reduction (DR): *Understanding and controlling reduction in molten salts, forming solid state materials cleanly and efficiently, specifically electroactive materials for enhanced electrochemical separation in the electrorefiner*
- Electrorefining & Speciation (ES): *Specific solid state materials production by dissolution and deposition of selected species with controlled composition and morphology, specifically ensuring proliferation resistance. Dissolution of stable materials, specifically Gen IV fuels (ceramic, nitride and carbide fuels) and production and characterisation of the MS soluble species;*
- Analysis (AN): *Establishment of molten salt analysis techniques, specifically the in-line sensing technology required for PR systems with modelling to understand molten salt materials processes.*

REFINE Research Consortium
A New Research Initiative for Sustainable Nuclear Fuel Reprocessing

The University of Nottingham
UNITED KINGDOM · CHINA · MALAYSIA

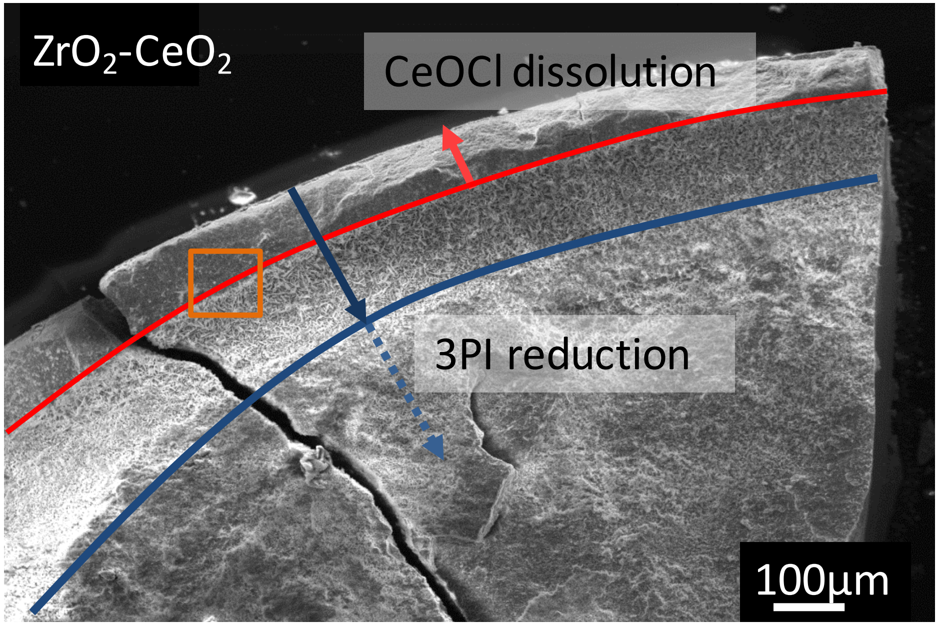
Direct reduction



Before reduction After reduction

Partitioning of TiO_2-CeO_2

Seems familiar...



Surface rich in **ZrO_2** & CeO_2

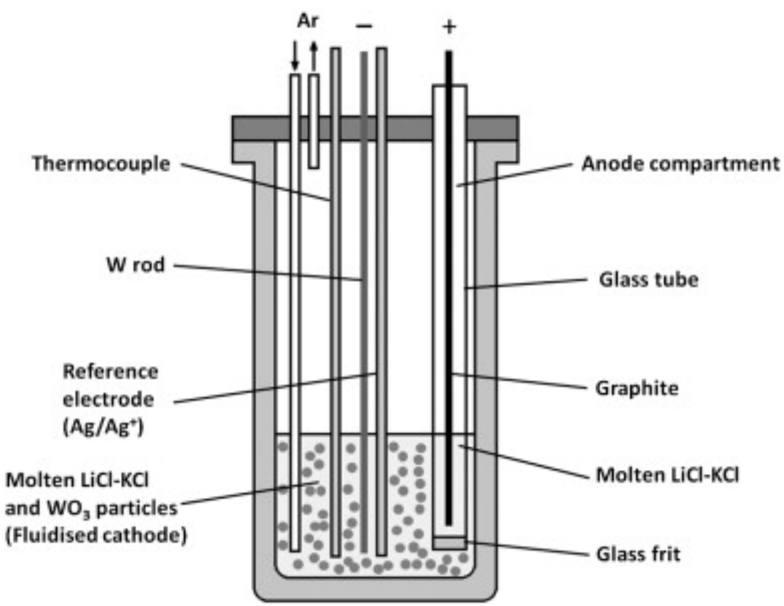
- CeOCl formation and dissolution rate limited (Cl^- ion coalescence)
- Surface readily undergoes partitioning.
- CeOCl solubility in LKE?

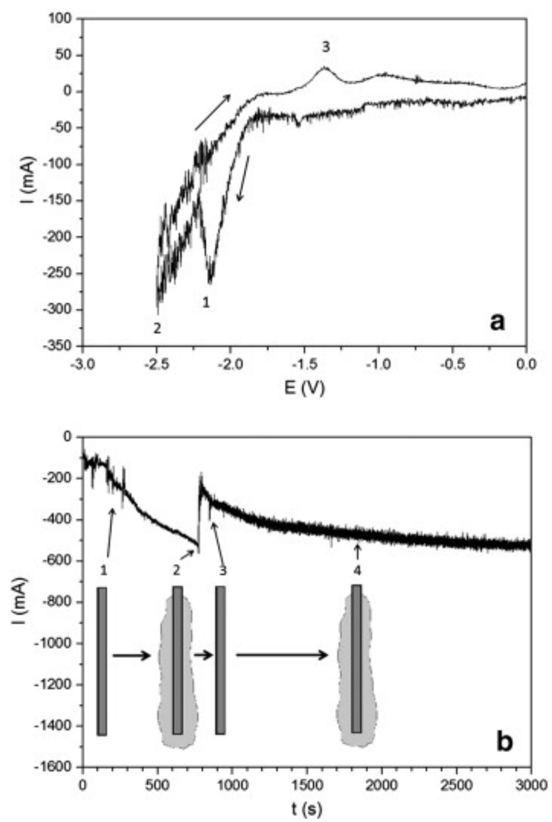
No Zr depletion because:

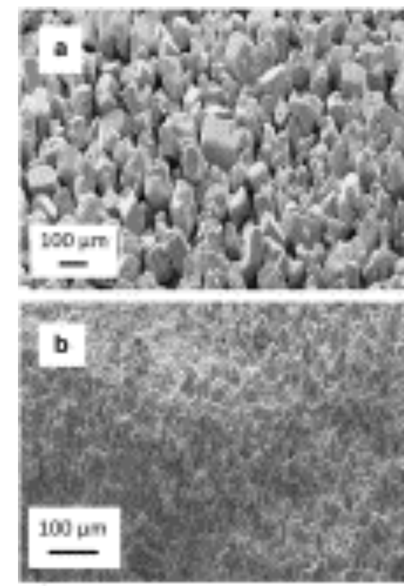
- E not sufficiently negative
- **No selective anodic dissolution**

DR Highlights

- Development of a fluidised bed reactor



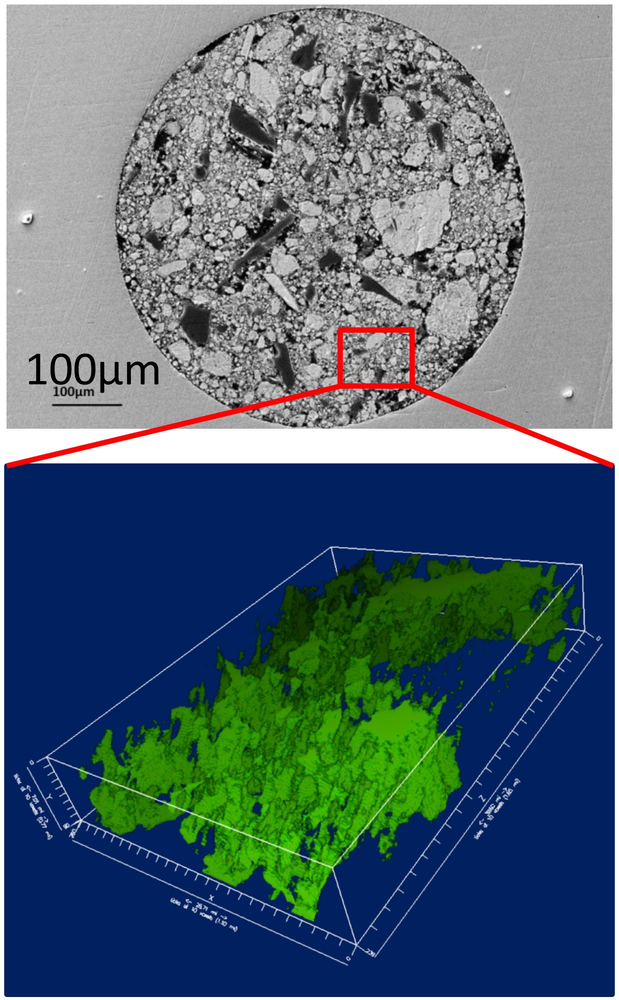




Adulaziz. R, Brown. L, Inman. D, Shearing. P & Brett D. (2014) Electrochemistry Communications

DR – Tomography Studies

UO_2



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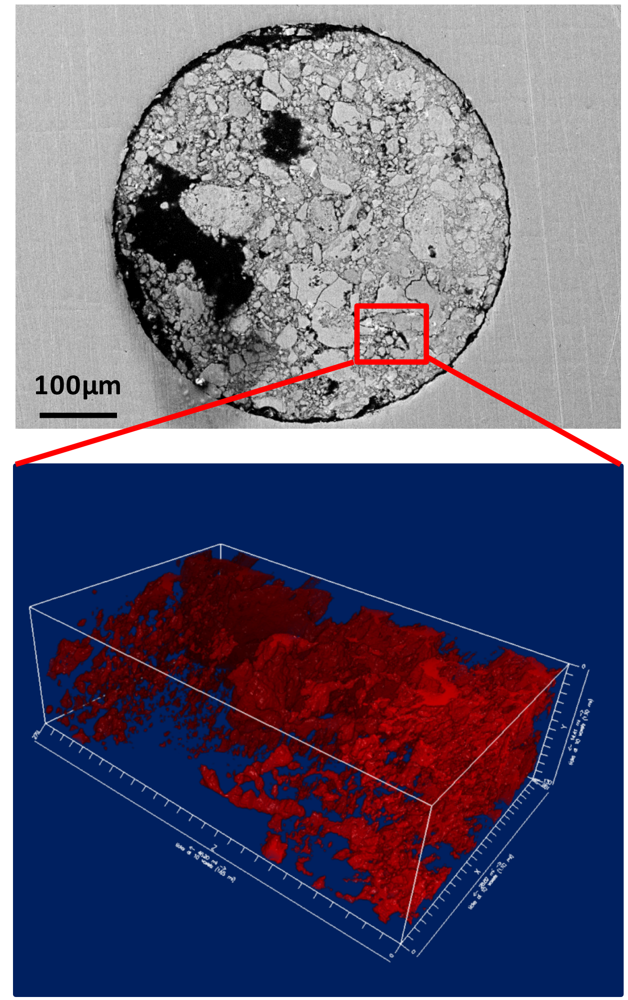
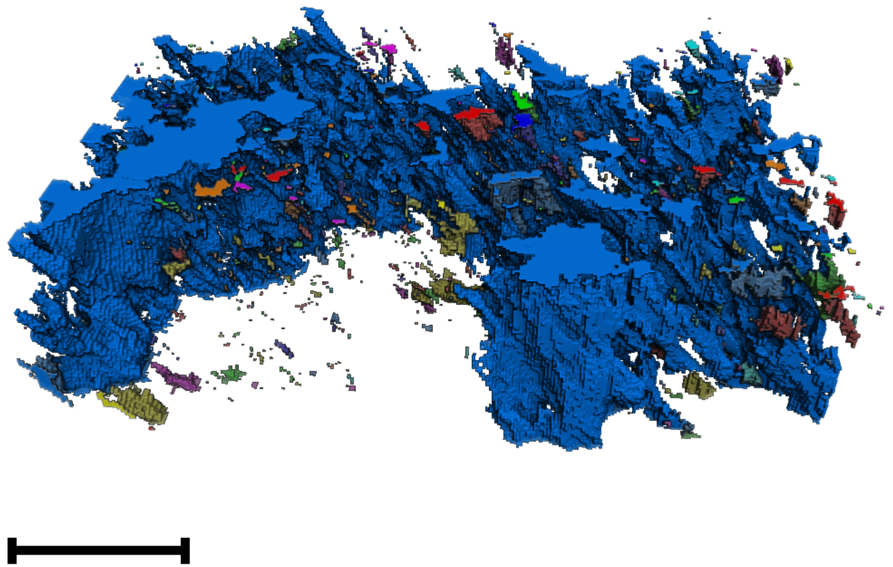


Tomography

UO_2



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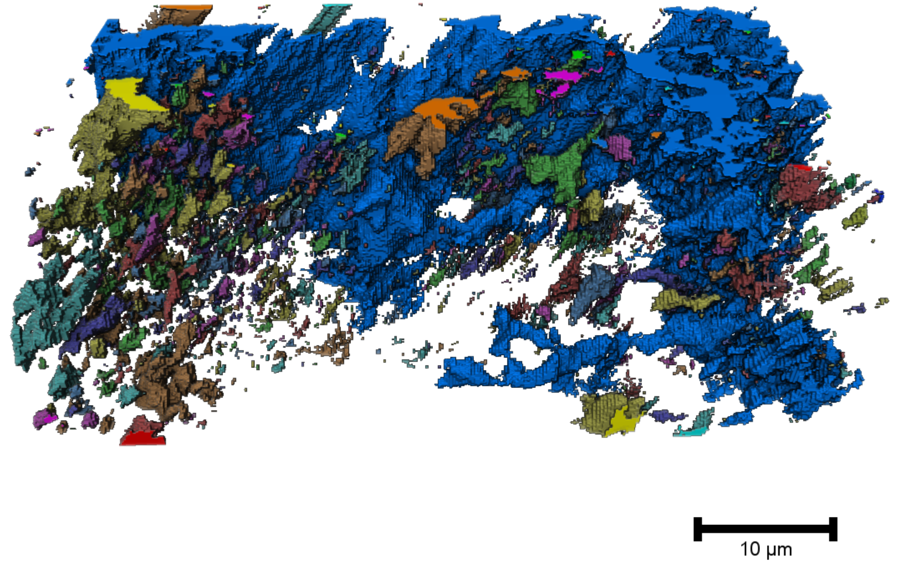
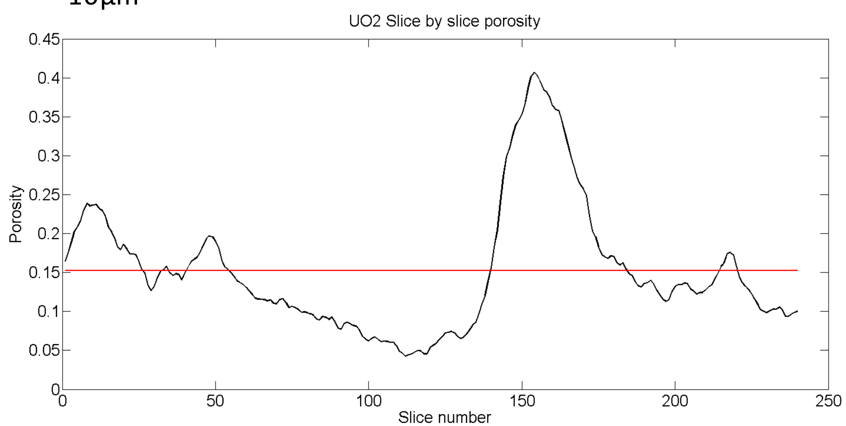
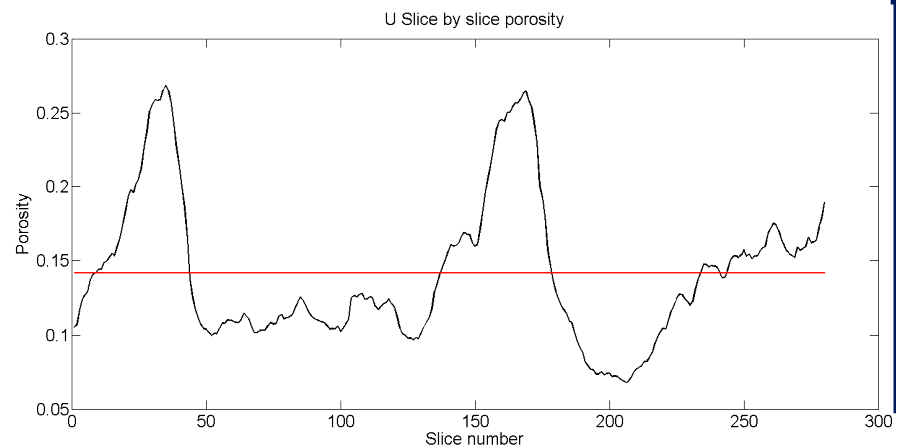
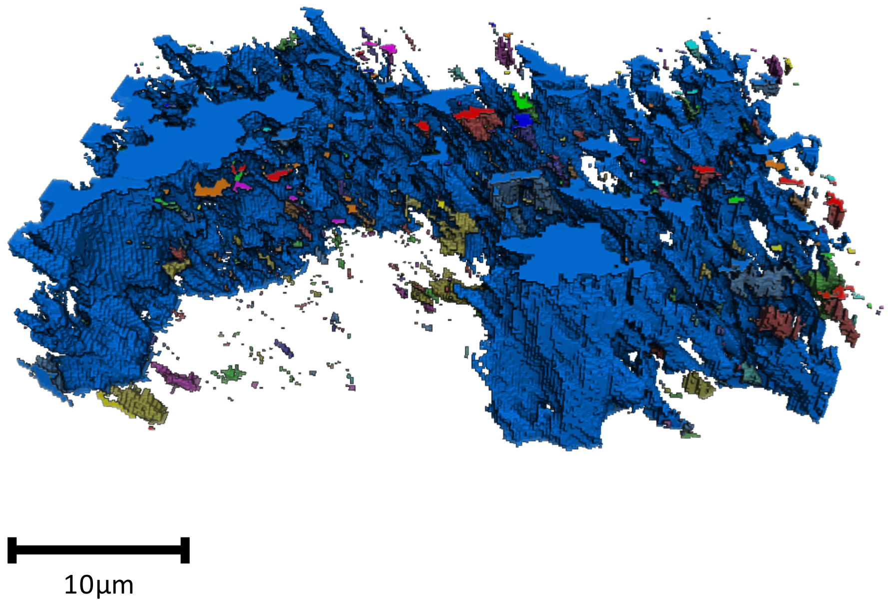


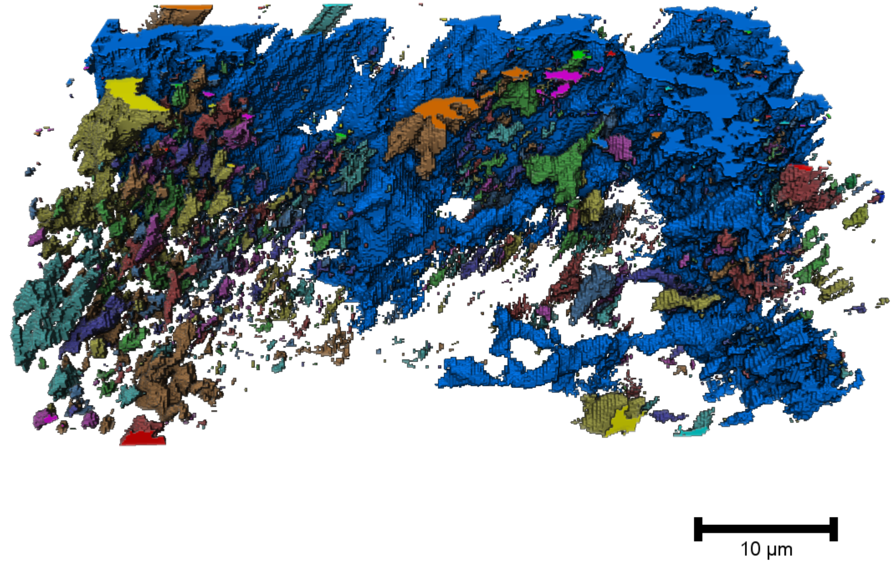




Tomography

UO_2	U

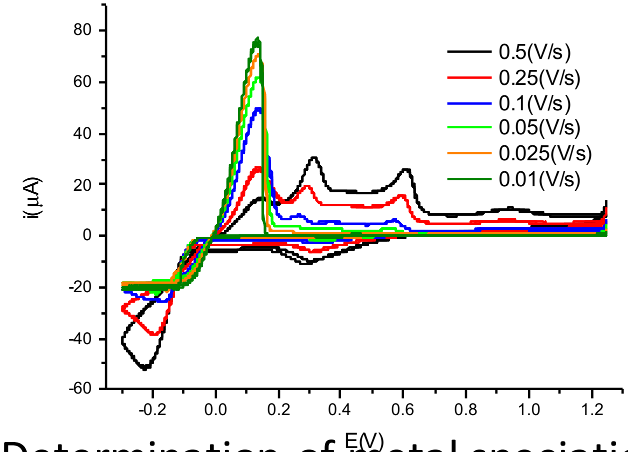




Number of Pores – 980	Number of Pores – 2695
Pore Connectivity – 97.7%	Pore Connectivity – 88.27%

ER Highlights

Liquid and Solid Cathode: fundamental electrorefining studies and characteristics



Powder XRD for quenched melt analyses

Some metal carbides can be converted to the oxide in molten salts (e.g. Cr_3C_2)

- Determination of metal speciation of carbide material (cf. UC fuel) in chloride melts to gain understanding of the behaviour of the metal:
  - Characterization of inert matrix fuel candidates and coated fuel surrogate material.
  - Elucidation of the fate of carbon in chloride melts from the chemical and anodic dissolution of carbide material.
  - Development of spectroscopic and analytical techniques for the identification of species formed during the chemical and anodic dissolution of UC in chloride melts.

AN highlights

The ability to monitor the salt composition in real time is of paramount importance for process control.

Current techniques

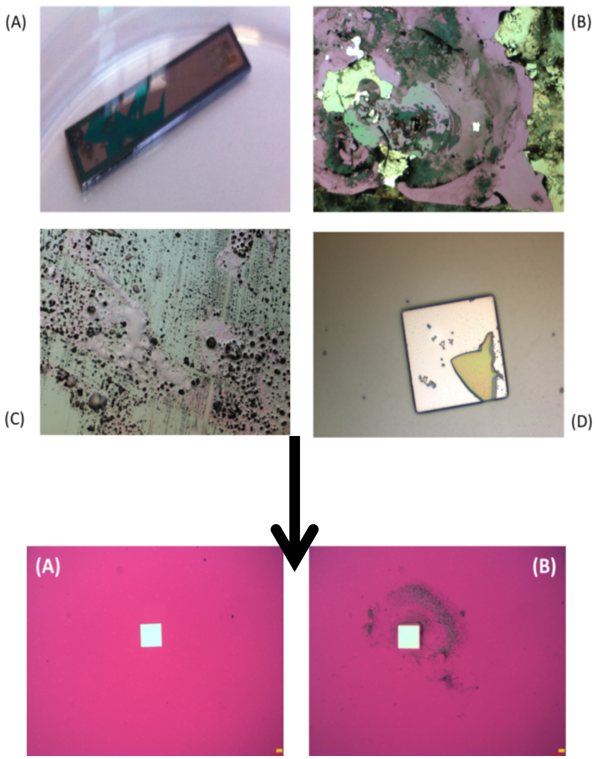
Ex situ techniques

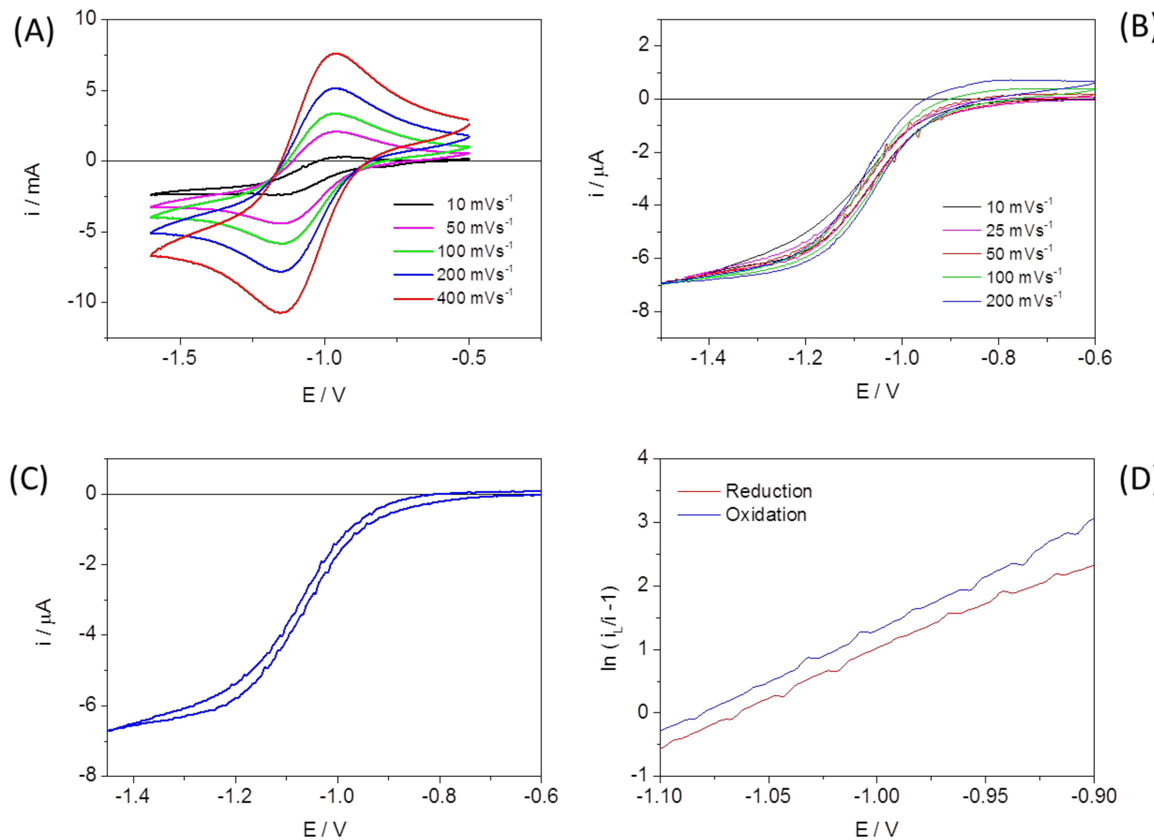
- HPLC analysis of recovered salt

In situ techniques

- Spectrophotmetric monitoring
- Electroanalysis
- Laser induced breakdown spectroscopy
- Off gas measurements
- Differential scanning calorimetry
- NMR?

Electrochemical device fabrication and analysis



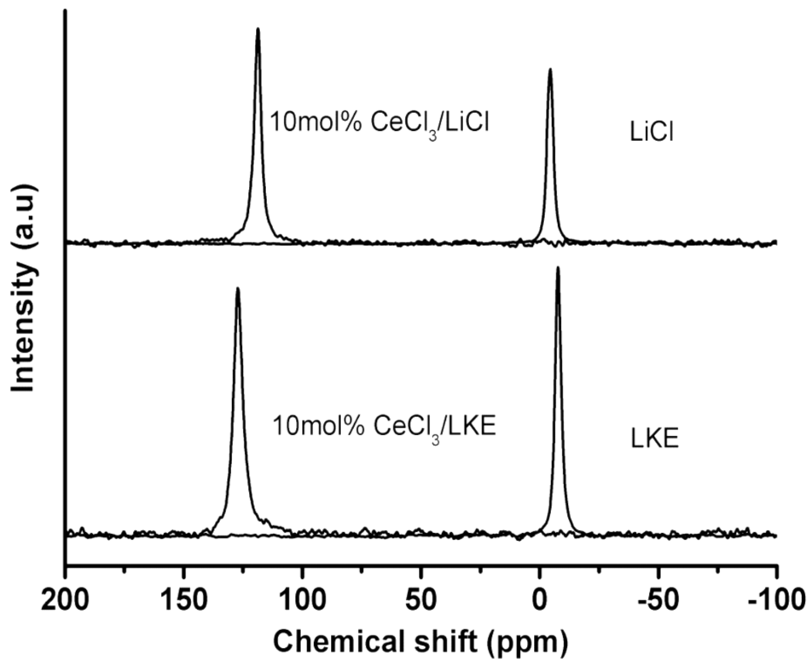


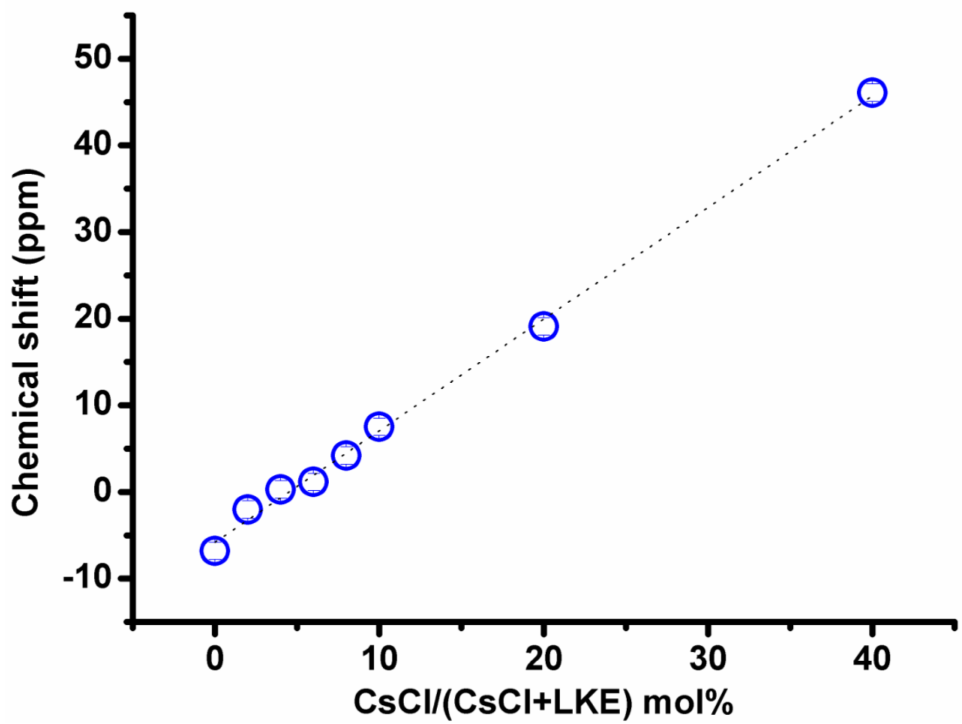
Corrigan, D.K., et al (2014) – Analytical Chemistry

NMR

- Fundamental properties of molten salts and solutes
- Development for in-line analysis in molten salt systems

$CeCl_3$/LKE





Effect of $CeCl_3$ on ^{35}Cl chemical shift: shielding of the ^{35}Cl, indicate stronger interaction between Cl^- and cations.

Compositional effect of additions of alkali chlorides (e.g. CsCl) and surrogates of Uranium and Plutonium (e.g. $CeCl_3$) on chemical shift of ^{35}Cl can are being studied by NMR.

Potential for monitoring concentrations during the electrorefining process.

Two active measurements at NNL

- UoN – Anthony Stevenson – electroreduction studies with PuO_2.
- Scheduled for Oct 2015.

- UoE – Dr Damion Corrigan – electrochemical measurement of Pu in LKE using molten salt compatible microelectrodes.
- Scheduled for Nov 2015.

- Led by – Dr Mike Harrison (NNL)

Summary

- The REFINE consortium is:
  - delivering essential molten salt (pyrochemical)
    - fundamentals
    - platform technologies
    - underpinning process development
    - training
  - for safe, dependable and sustainable UK nuclear fuel reprocessing
- Present focus: outputs, engagement, next steps….
  - Outreach meeting: Royal Acad of Engineering, end 2015
  - 8 journal publications to date, 20 by programme end
  - 4 keynote presentations at international conferences
  - www.refine.eng.ed.ac.uk Email: a.mount@ed.ac.uk

Pyroprocessing Research Laboratory

4. Pyrochemical reprocessing laboratory

4.1 Introduction

DECC pyrochemical reprocessing laboratory to develop and demonstrate integrated pyrochemical reprocessing of nuclear fuel using fuel-relevant compositional mixtures, and to develop the required process monitoring, at laboratory scale.

4.3 Capability requirements

The laboratory is expected to consist of a suite of interconnected integrated controlled-atmosphere dry-boxes, equipped with the necessary furnaces and cell systems required for pyrochemical measurement of each of the essential elements of the process at the laboratory scale. There should be internal connectivity between these elements to enable the integrated process development required to establish and demonstrate the complete fuel recycle process.

Electrochemical and spectroscopic characterisation equipment will be needed for fundamental characterisation, and to further develop the process monitoring and characterisation methods developed in REFINE and EU programmes.

Operating principles and research objectives of the PRL

- Development of a complete pyroprocessing flow sheet
- A system of dry boxes to enable research in all aspects of pyroprocessing
- Four interconnected dry boxes for:
  - Salt Preparation
  - Static studies on electro-reduction and refining
  - Dynamic studies on electro-refining and high temperature online monitoring
  - Ambient temperature analyses of process samples
- LKE and $CaCl_2$ capability
- Interconnected with "T" junctions for optimum booking flexibility
- Designed for work with surrogates, fuel simulants (e.g. TRISO particles and carbides) and depleted Uranium
- Common optical and electrical feed through for connection of potentiostats and optical spectrometers
- Fluoride capability
- Will open March 2016……….