Corrosion and Leaching of Carbide Fuels in a Geological Disposal Facility setting.

Dimitris Samaras, University of Bristol

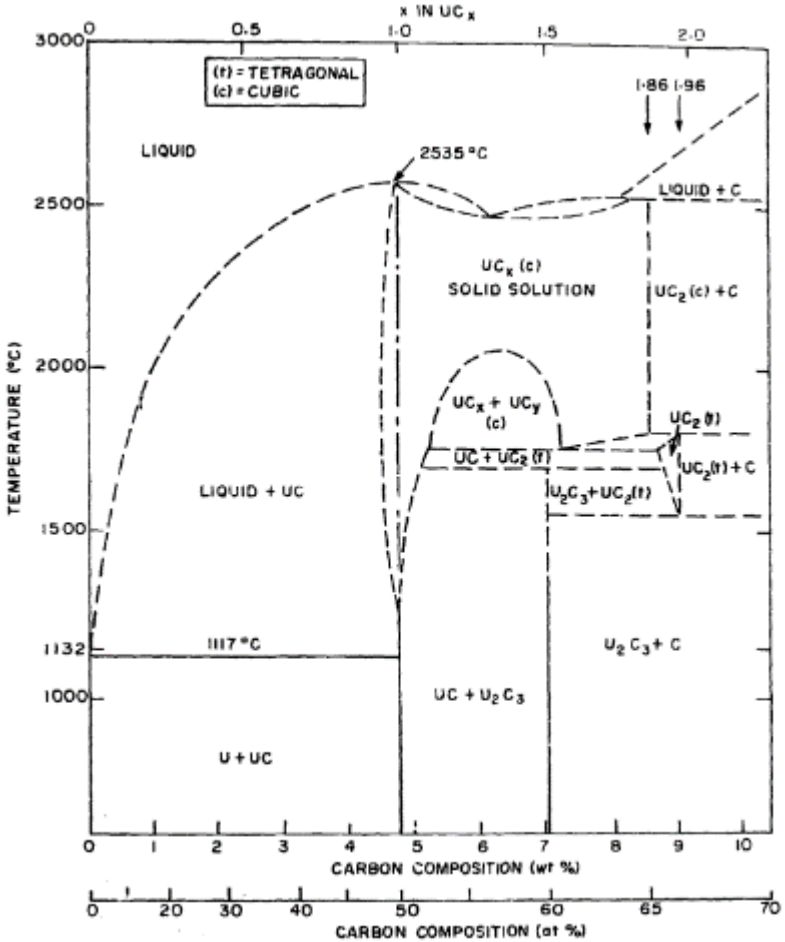
bristol.ac.uk

Intro

- NDA UC Inventory
- Advantages
  - Good dimensional stability
  - Fission Gas Retention
  - 30% higher density over UO_2
  - 6x Thermal Conductivity over UO_2

Stoichiometries

- 3 stoichiometries
- UC most preferred
  - UC_2 : BCT < - > FCC
  - U_2C_3 -> UC_2 + UC



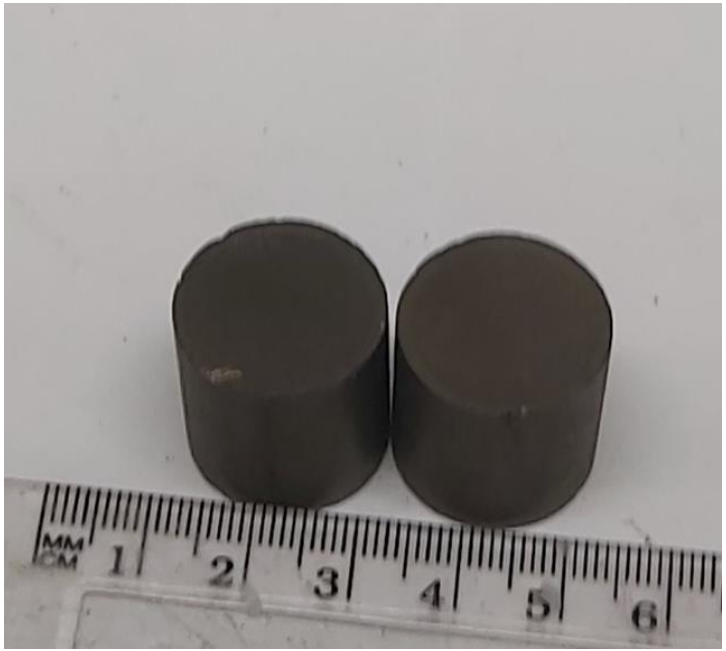
Uranium-Carbon Phase Diagram
Image: Jones, R. W. (1972). *Uranium Carbide as a Nuclear Fuel*.

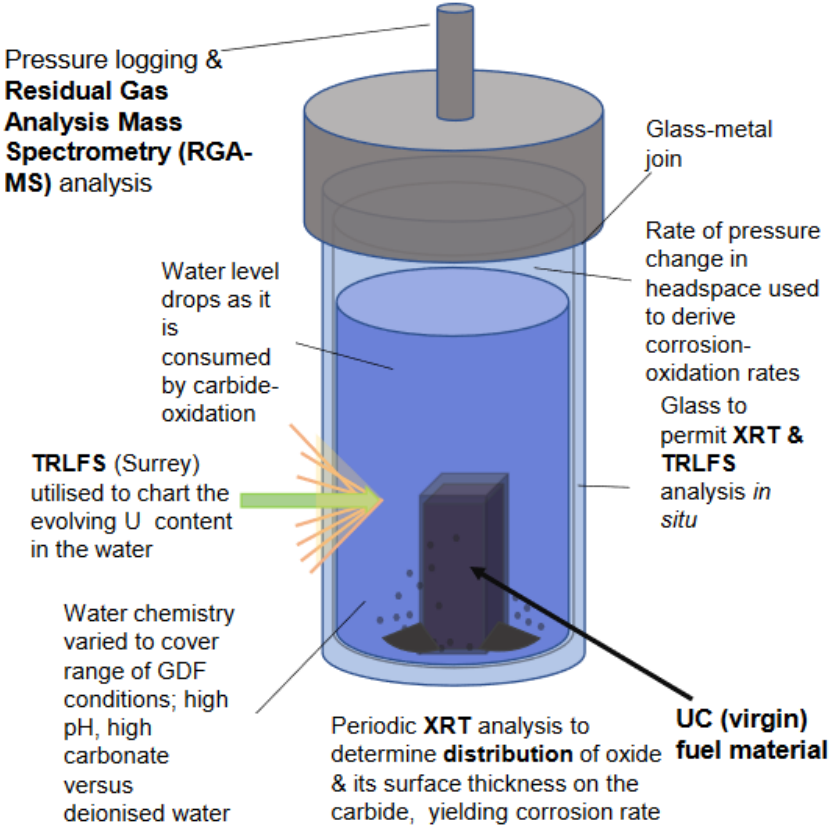
Hydrolysis - Oxidation

- Rate dependant on temperature
  - Mild at 20°C ; vigorous at around and above 40 °C
    K.M. Taylor and C.H. McMurtry. Synthesis and Fabrication of Refractory Uranium Compounds, *U.S. Atomic Energy Commission,* 1960
- Product formulas dependant on the stoichiometry
  M.J Bradley and L.M. Ferris , Hydrolysis of Uranium Carbides between 25 and 100 °C : I and II (1962 & 1964)
  - UC -> mainly CH_4
  - U_2C_3, UC_2 & mixtures -> mainly H_2, C_2H_6, C_3H_8, and heavier hydrocarbons
- Diluted in groundwater
  - Excavation and waste deposition
- Dry oxidation: linear rate law,
  K.A. Peakall and J.E. Antill, Oxidation of Uranium Monocarbide, *Journal of the Less-Common Materials,* 1962
  - N.B. Min temp: 230°C

The samples

- O_2/N_2-free environment
  - Ar glovebox
- 6 MBq
- Results
  - 2 cm ø x 2 cm height
  - Weight: 82 gr each



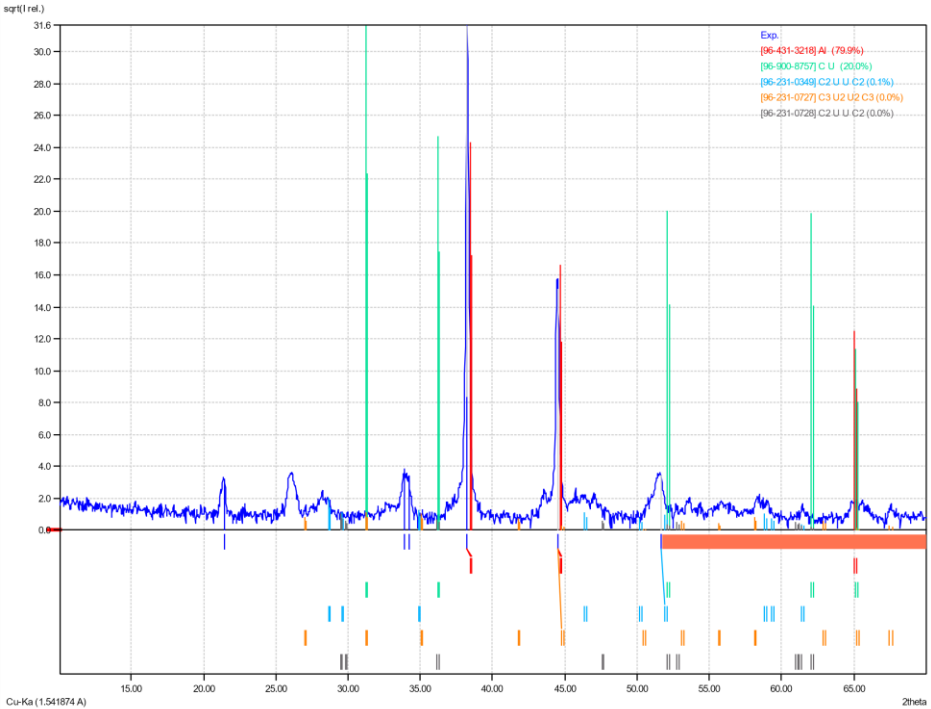


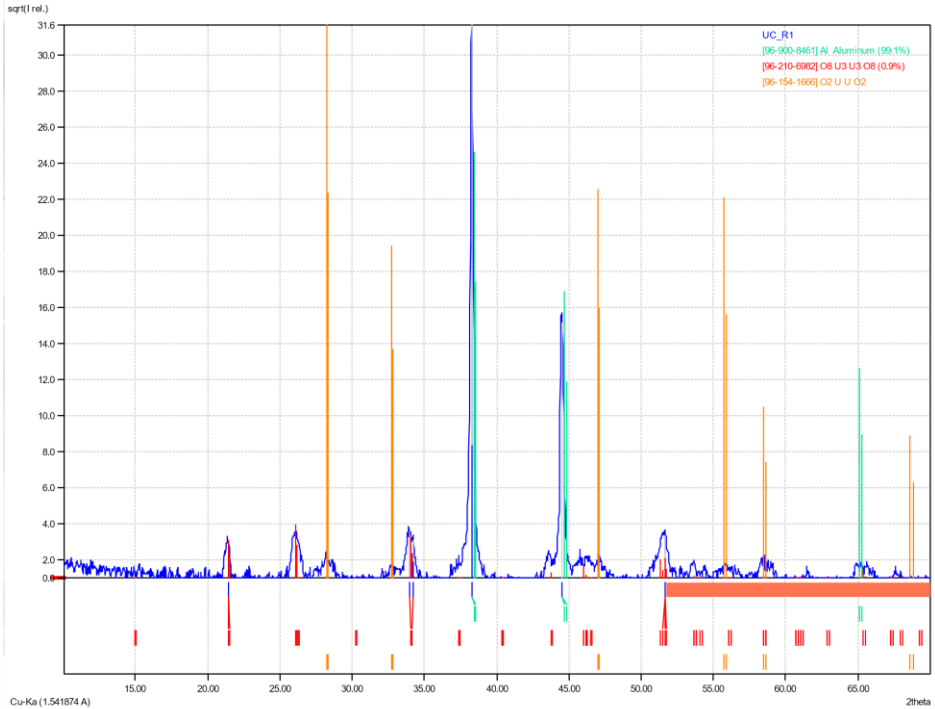
- Needed to cut into 0.5cm-sized cubes
- Accutom
- However…

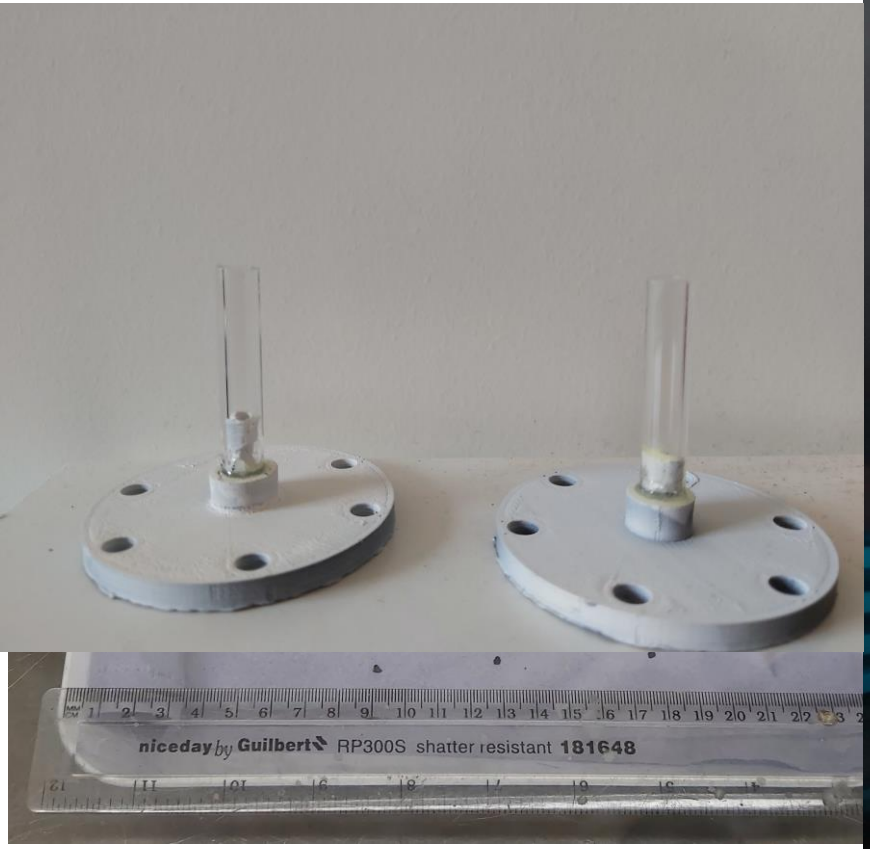
UC Corrosion Experimental Cell Concept

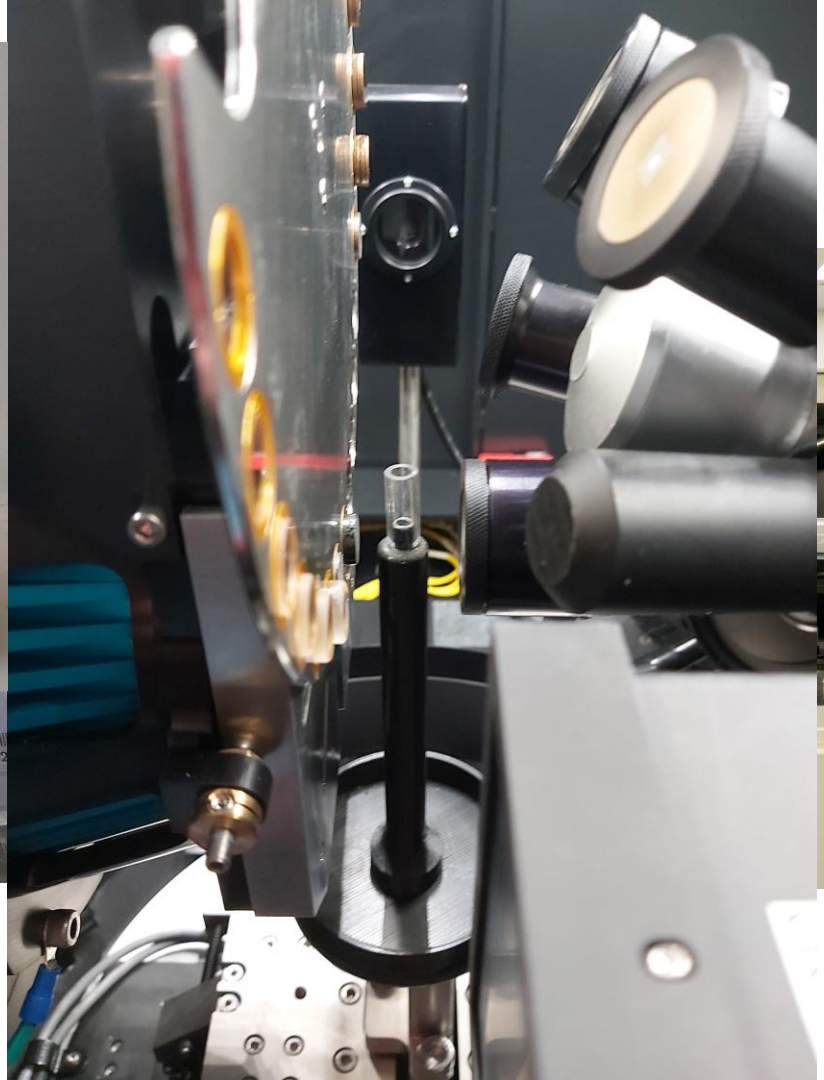
Image: Prof. David Reed, TRANSCEND 1st Industry Roadshow, Warrington

…t…


sqrt(I rel.)
Exp.
[96-431-3218] Al (79.9%)
[96-900-8757] C U (20.0%)
[96-231-0349] C2 U U C2 (0.1%)
[96-231-0727] C3 U2 U2 C3 (0.0%)
[96-231-0728] C2 U U C2 (0.0%)
Cu-Ka (1.541874 A)
2theta


sqrt(I rel.)
UC_R1
[96-900-8461] Al Aluminum (99.1%)
[96-210-6982] O8 U3 U3 O8 (0.9%)
[96-154-1666] O2 U U O2
Cu-Ka (1.541874 A)
2theta

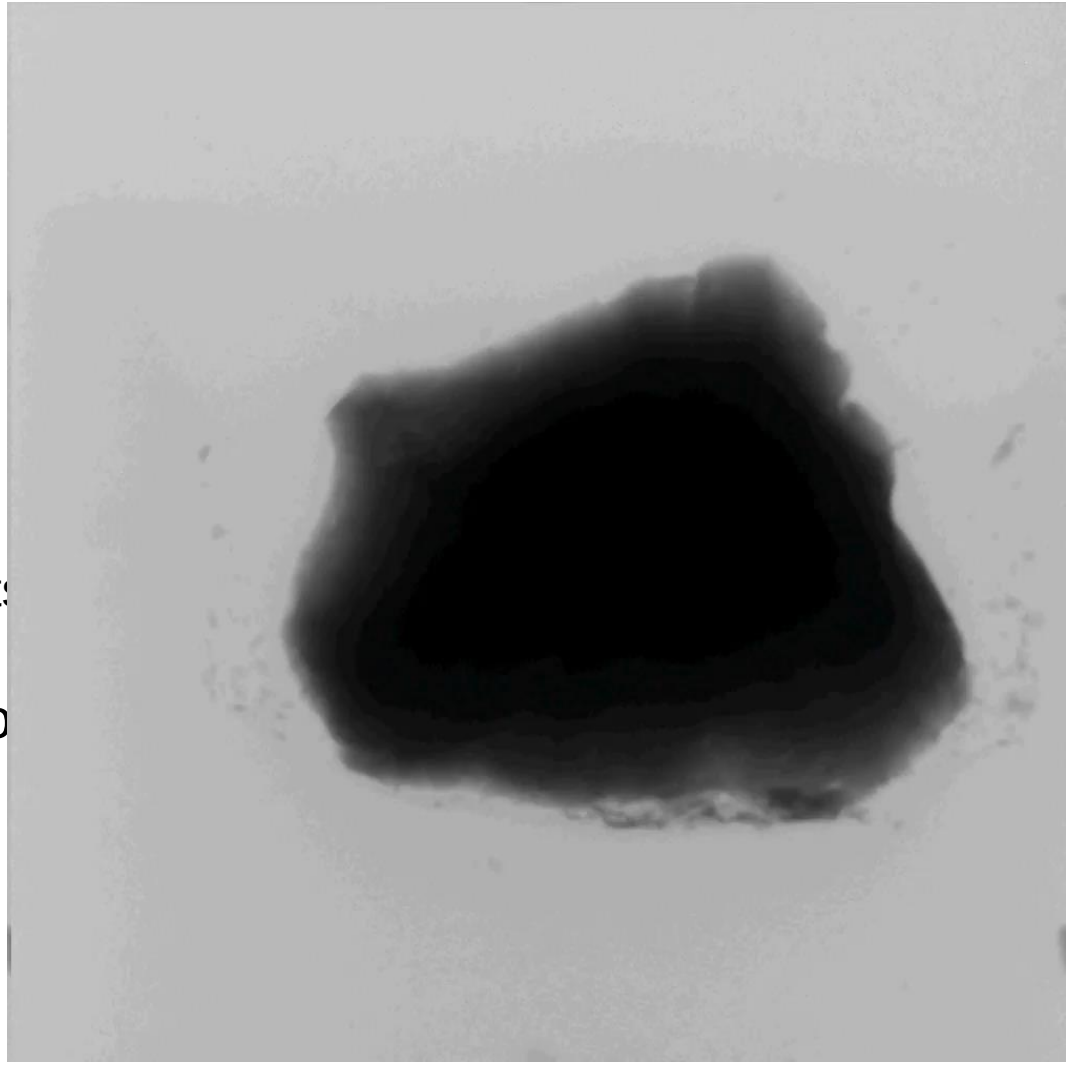




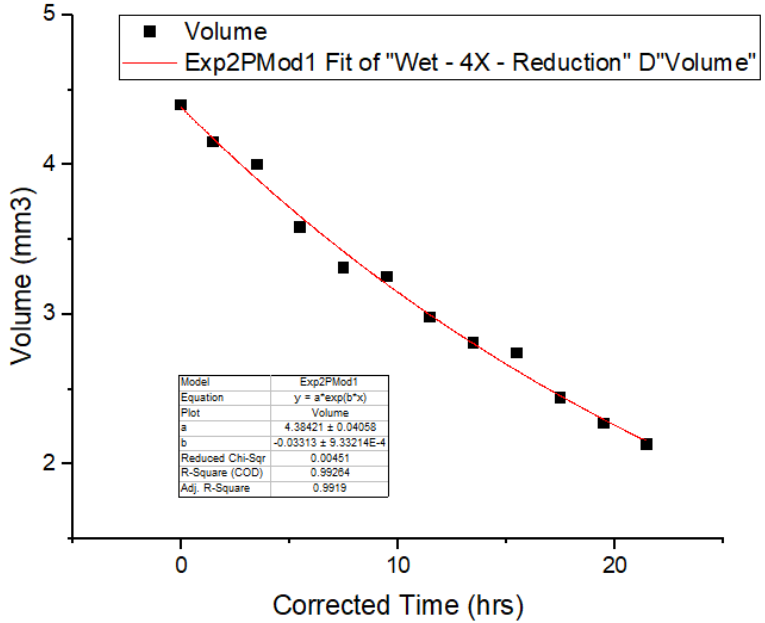
bristol.ac.uk

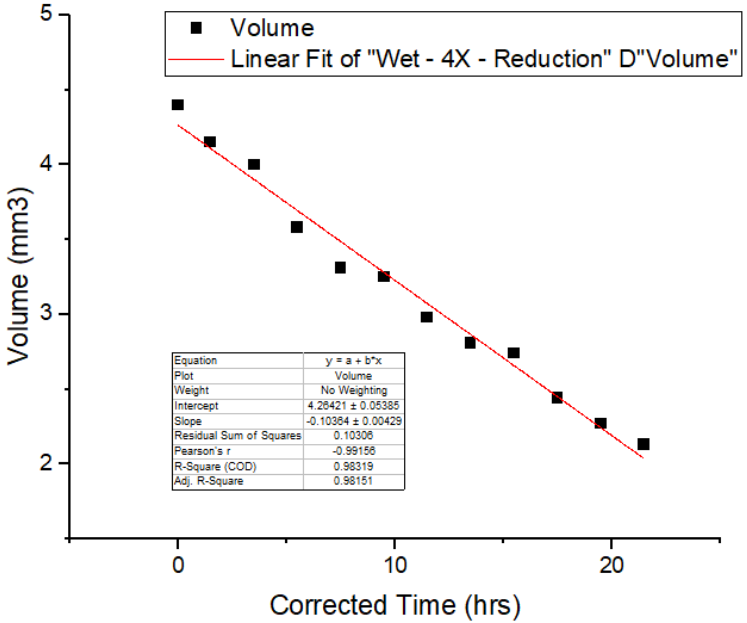
X-Ray Tomography

- Two-step process
- Dry Scan -> Suitable Magnification Scale
  - 0.4X, 4X, 10X, 20X, 40X
  - 4X for millimetre-size fragments
    0.4X for bigger ones
- Wet Scan to observe corrosio



Data Analysis





Limitations so Far

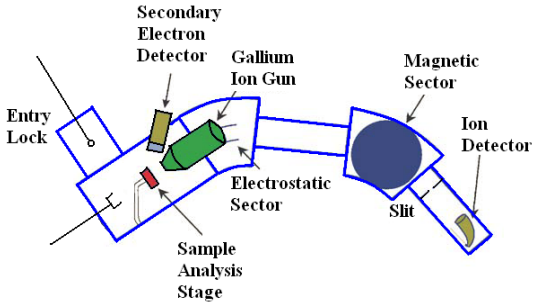
- Small water reservoir - Corrosion Halt
  - Larger glass tube attachable/detachable to a plastic base
- Vibration
  - More secure fixing; vertical placement at all times
- No pre-corrosion composition identification
  - X-Ray Diffraction on each fragment pre- and post-hydrolysis

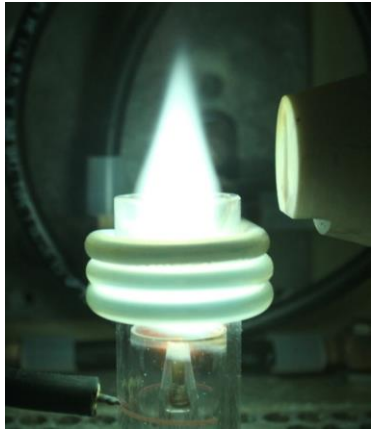
Future work

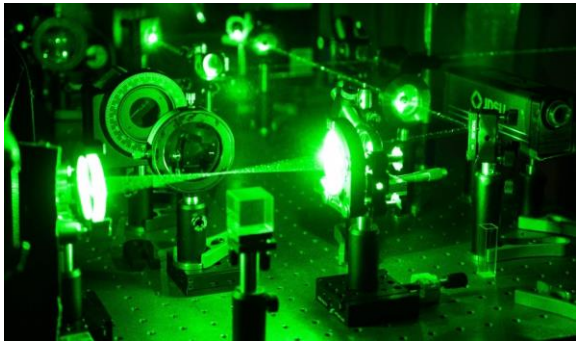
- Additional X-Ray experiments
  - Tomography
  - Diffraction
  - Synchrotron
- High-Speed Atomic Force Microscopy
  - Nanoscale

Future Work

- Intact Pellet - partial corrosion
  - EM
  - SIMS
- Solution Analysis
  - TRLFS – University of Surrey
  - ICP-MS/OES







Thank you for listening!

Acknowledgements

- Professor Tom Scott
- Dr. Ross Springell
- Dr. Rosie Hibberd
- Dr. Christopher Jones
- Dr. Jacek Wasik

This research was funded by AN INVESTIGATION INTO CORROSION AND LEACHING OF CARBIDE FUELS IN A GEOLOGICAL DISPOSAL FACILITY (GDF) SETTING, project code RWM232

bristol.ac.uk