Diamond, Decommissioning and Disposal of the UK's Nuclear Legacy

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Active Materials Building Launch March 1st, 2022

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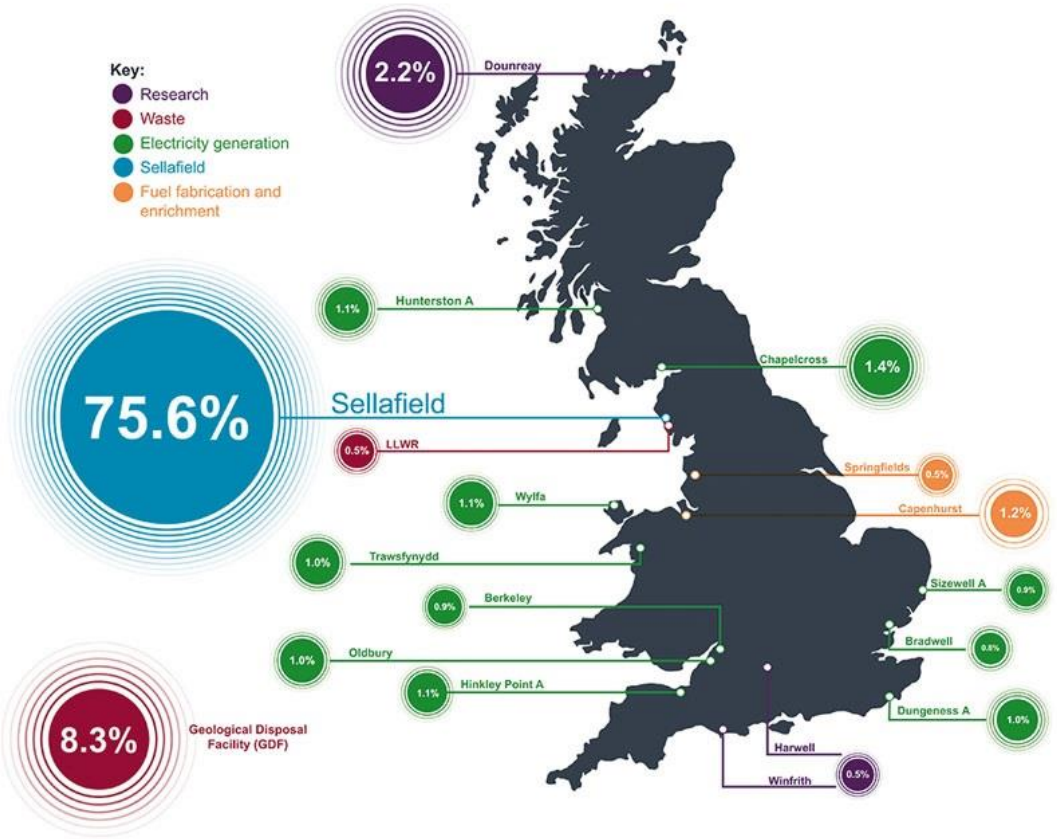
Talk Structure

- The scale of the challenges, the “nuclear legacy”
- Examples of direct impact
  - Sellafield – EARP plant
  - Sellafield – SIXEP plant
- NNUF RADER Facility UoM
- A prospect for the AMB and wider users

Britain's Nuclear Legacy

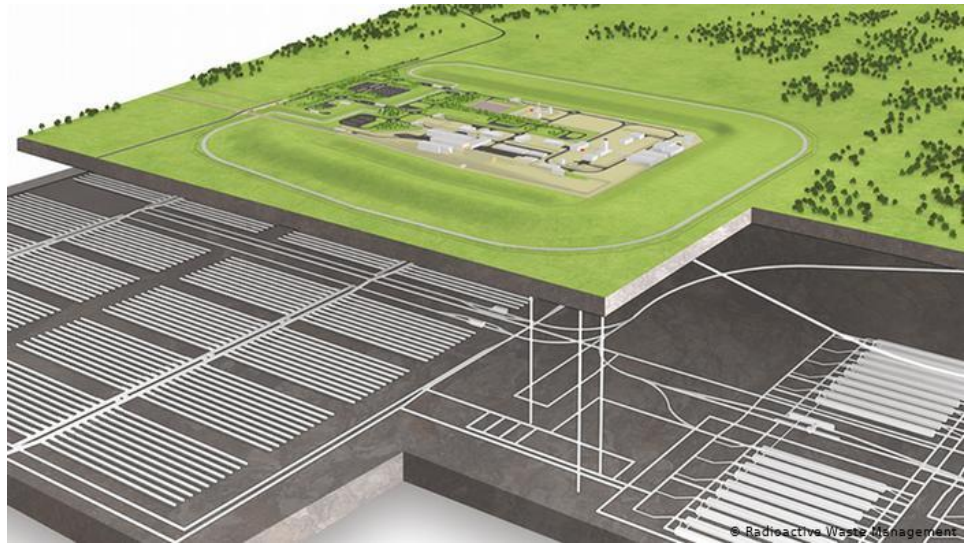
- Britain has historic nuclear sites which require clean-up
- Estimated cost and timescale
  - £126,000,000,000
  - >120 years
- Nuclear sector deal
  - minus 20 % challenge

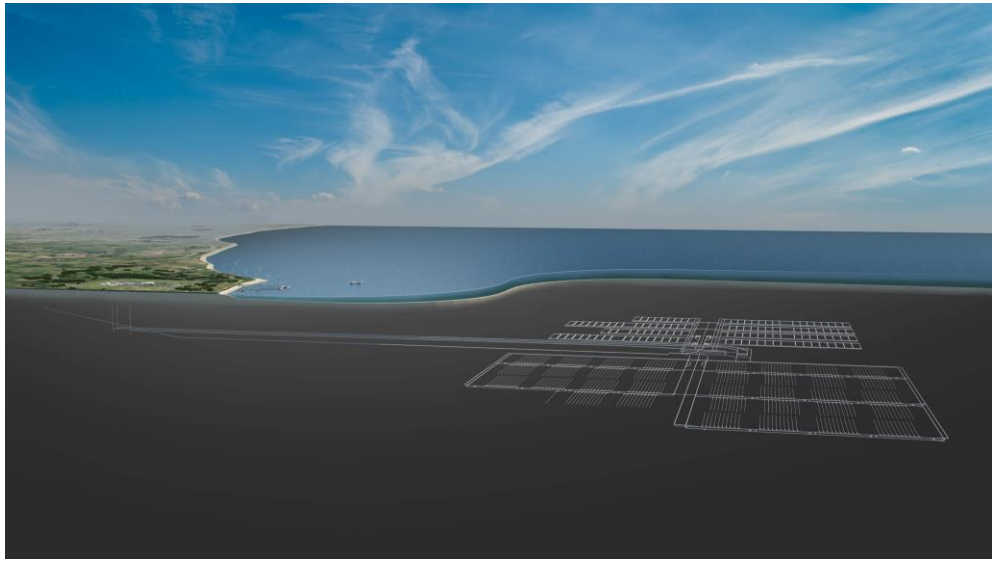


Nuclear provision: the cost of cleaning up Britain's historic nuclear sites. https://www.gov.uk/government/publications/nuclear-provision-explaining-the-cost-of-cleaning-up-britains-nuclear-legacy/nuclear-provision-explaining-the-cost-of-cleaning-up-britains-nuclear-legacy

Britain's Nuclear Legacy

- Legacy of higher activity radioactive wastes - 60+ years nuclear power / nuclear weapons
- 300,000 higher activity waste packages destined for deep geological disposal
- £20-50 billion pound, 100+ year project
- siting via volunteer communities happening now (2021/2) (west Cumbria x 3 community partnerships, Lincolnshire x 1 working group)



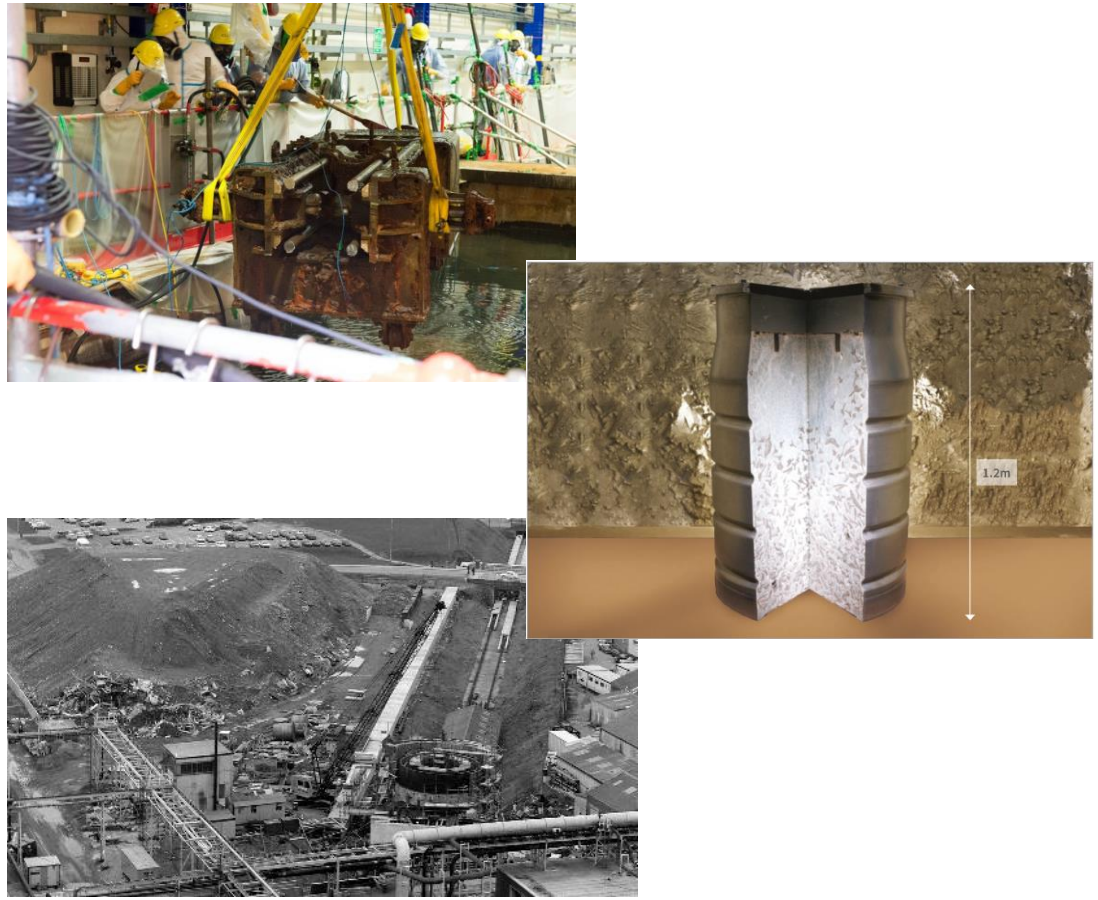


https://ukinventory.nda.gov.uk/the-2019-inventory/infographics/

Decommissioning, Disposal

- Fission products
- Actinides
- Activation products

- Understanding of, optimisation of, processes → implies mechanism



https://ukinventory.nda.gov.uk/wp-content/uploads/2020/01/2019-Waste-Report-Final.pdf

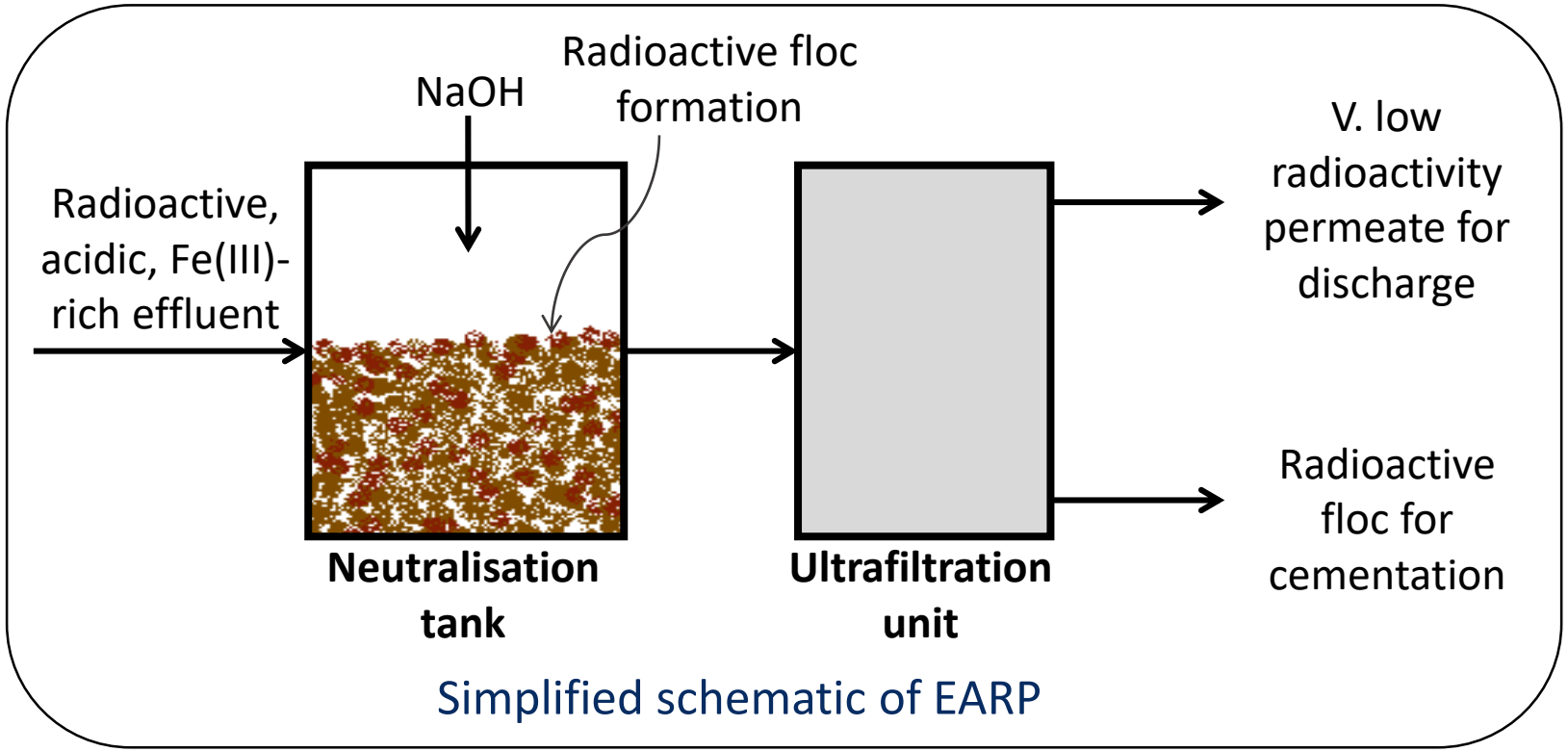
Decommissioning - Sellafield Effluent Treatment

- Sellafield Effluents
  - Acidic from reprocessing – Enhanced Actinide Treatment Plant (EARP)
  - Alkaline from Sellafield Legacy Ponds and Silos – Site Ion Exchange Plant (SIXEP)
- Effluent treatment underpins ongoing site decommissioning operations

https://nda.blog.gov.uk/2018/09/07/what-is-sellafield/
https://www.gov.uk/government/news/sellafield-ltd-to-restart-reprocessing-and-further-boost-operations-in-2020

EARP – Acidic Effluent, Ferrihydrite Precipitation

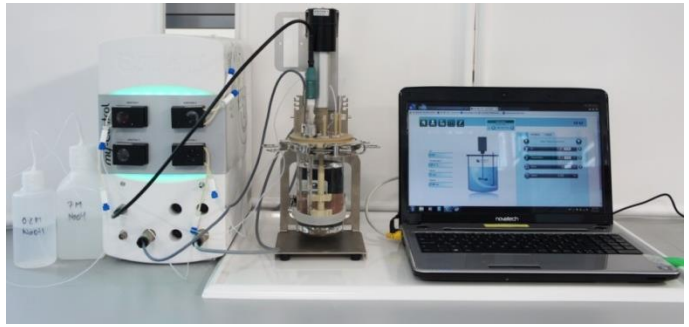
- Co-precipitates radionuclides with iron oxyhydroxide floc
- Two stages: **chemical neutralisation** followed by **physical separation**



Simplified schematic of EARP

EARP – Acidic Effluent, Ferrihydrite Precipitation

EARP – Acidic Effluent, Ferrihydrite Precipitation



Initial effluent:

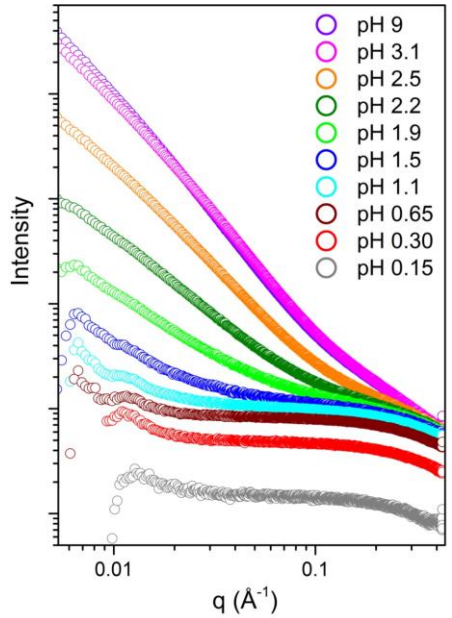
- 1M HNO_3
- 7.16mM $Fe(NO_3)_3$
- +/- Radionuclides

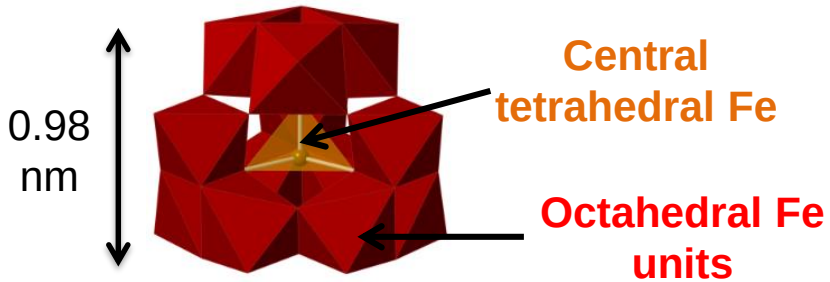
Final Product:

- Fe(III) oxyhydroxide solid
- +/- Radionuclides associated with the solid phase
- Cleaned effluent

pH 0.1 Sodium hydroxide addition **pH 9**

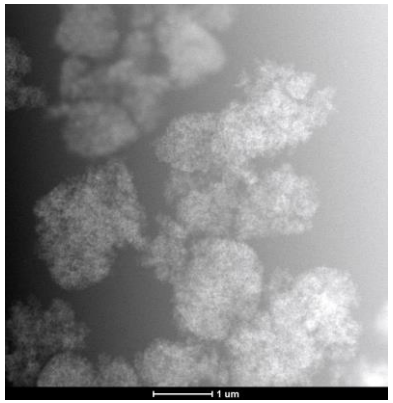
Ferrihydrite formation pathway in EARP

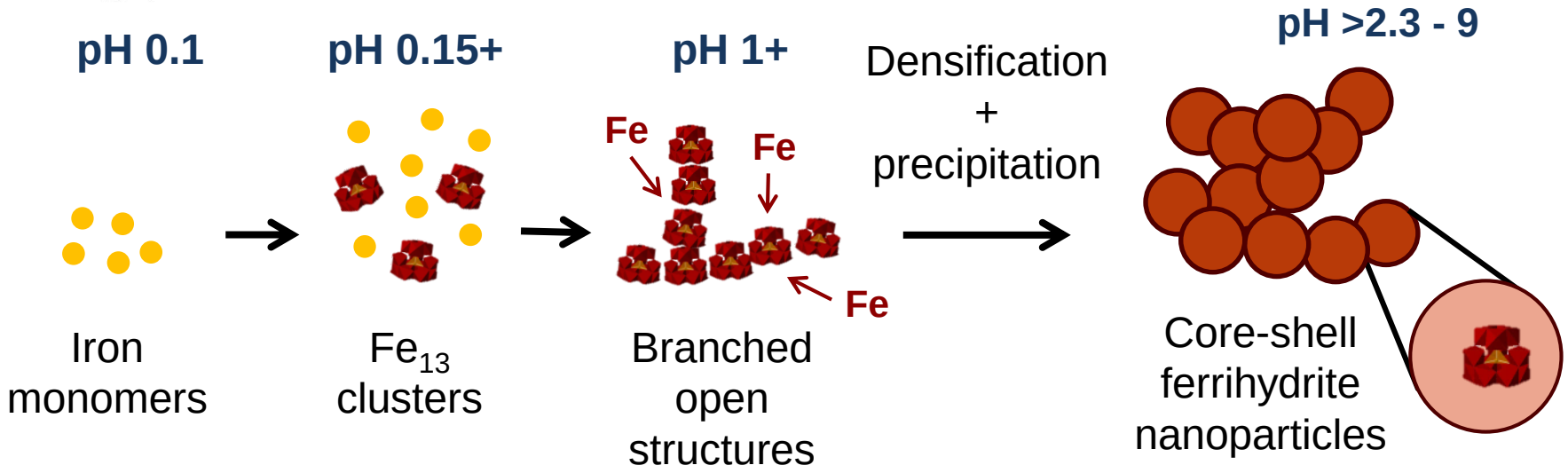




Fe_{13} Keggin cluster ($Fe_{13}O_{40}$)

In situ TEM pH 9

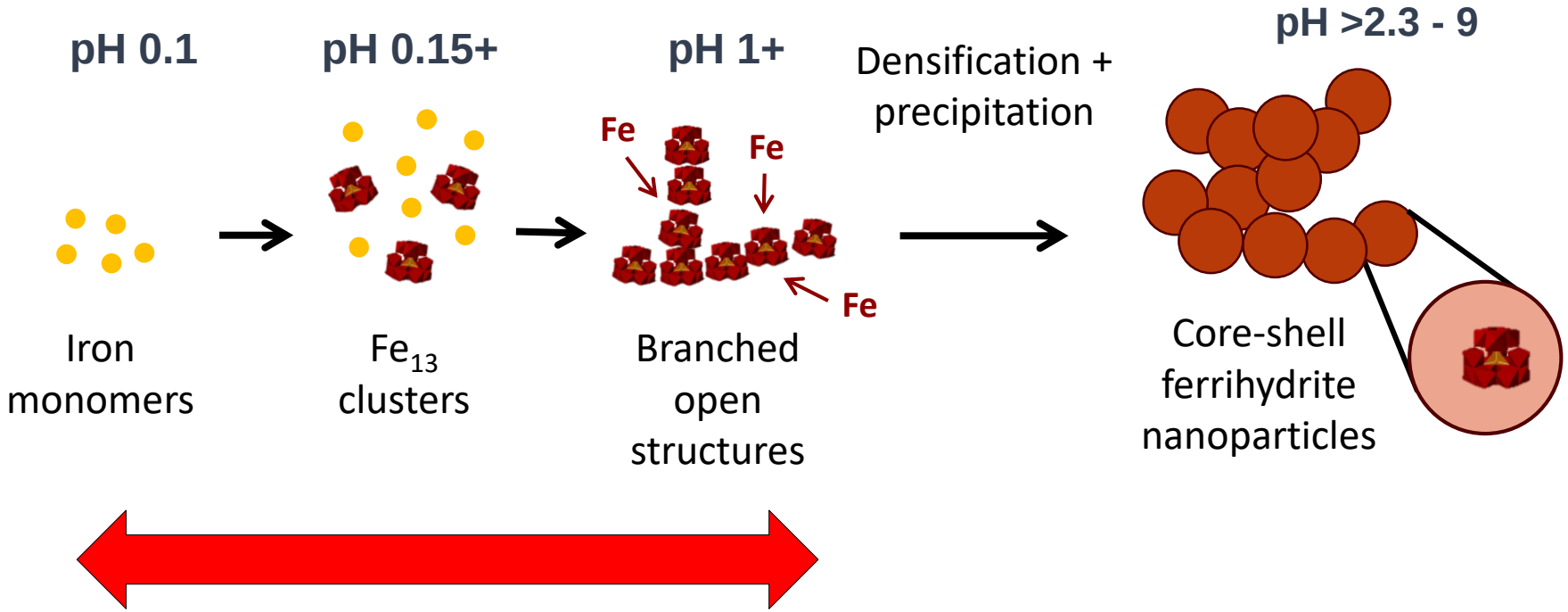




Sadeghi *et al.* (2015), Science, 347, 1359

Weatherill et al. (2016) Environmental Science and Technology 50, 9333–9342.

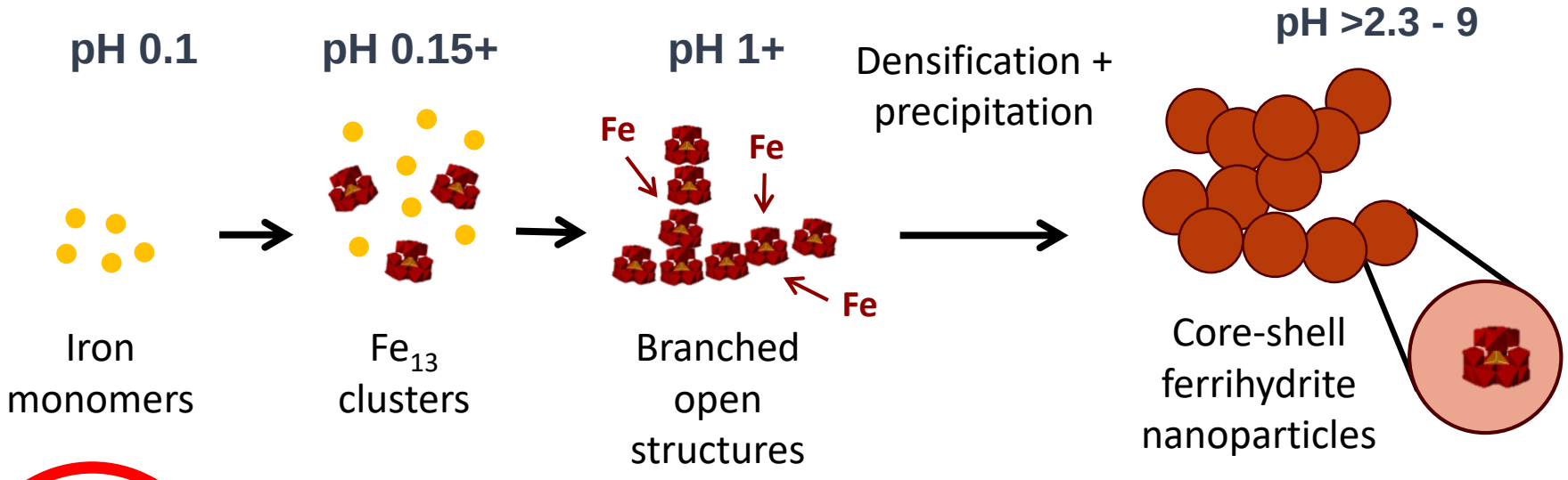
Informing Optimisation of EARP Operations

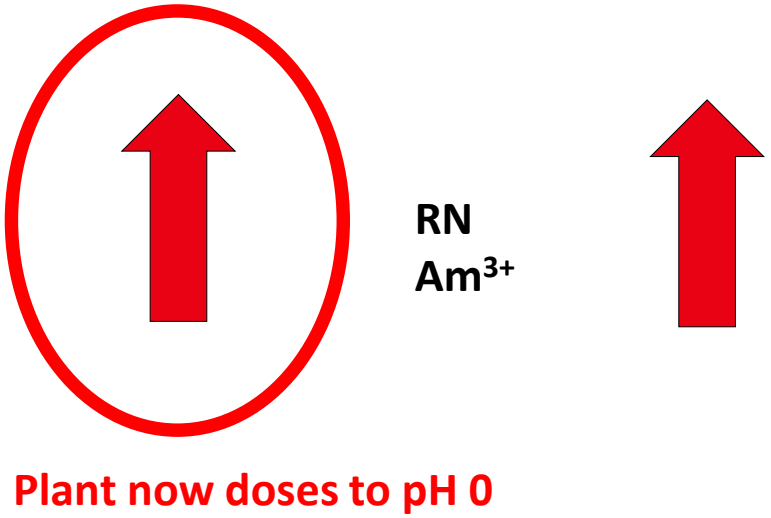


classic model – pH 0 ↔ 1.5 no difference as no precipitate

EARP influent range of pH

Informing Optimisation of EARP Operations





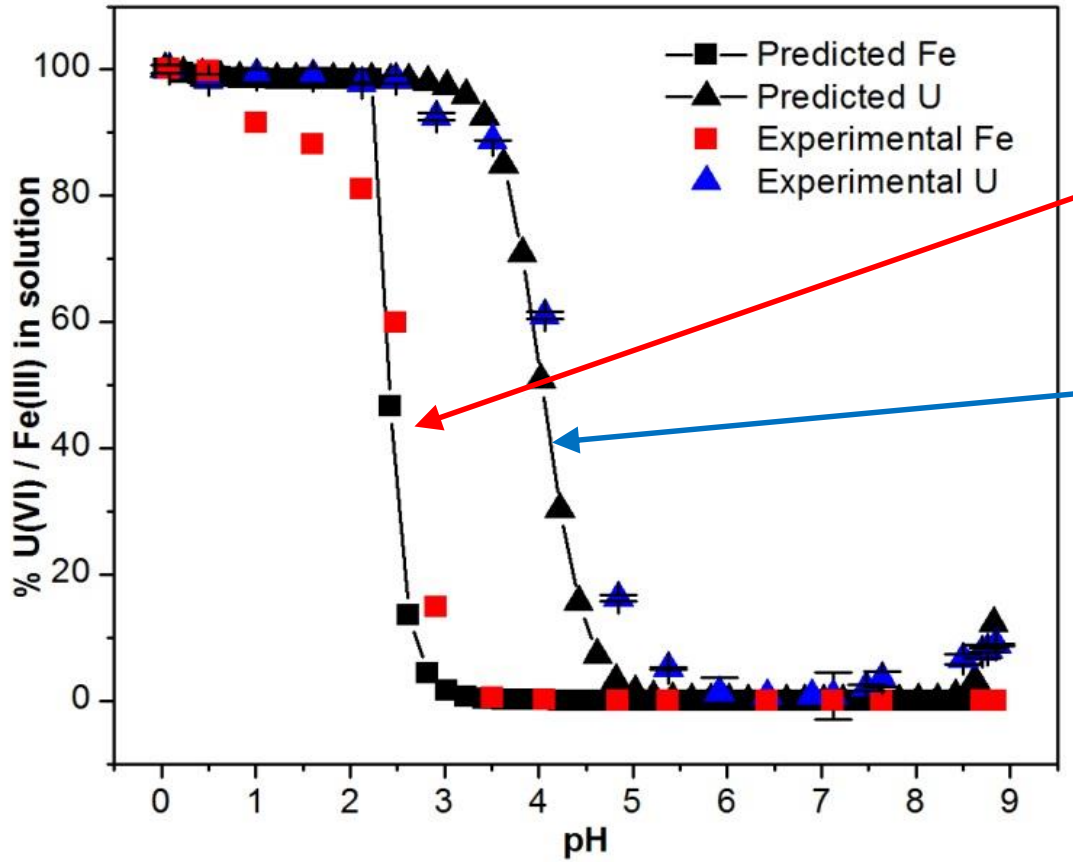
Plant now doses to pH 0

D_F improved at lower initial pH

NNL – Am^{3+} expt
Sellafield – historical data

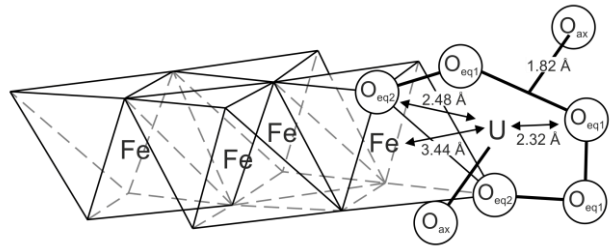
Further reduced discharges

Radionuclide Behaviour in EARP
Uranium uptake during ferrihydrite formation



Fe(oxyhydr)oxide ppt

U(VI)-sorption

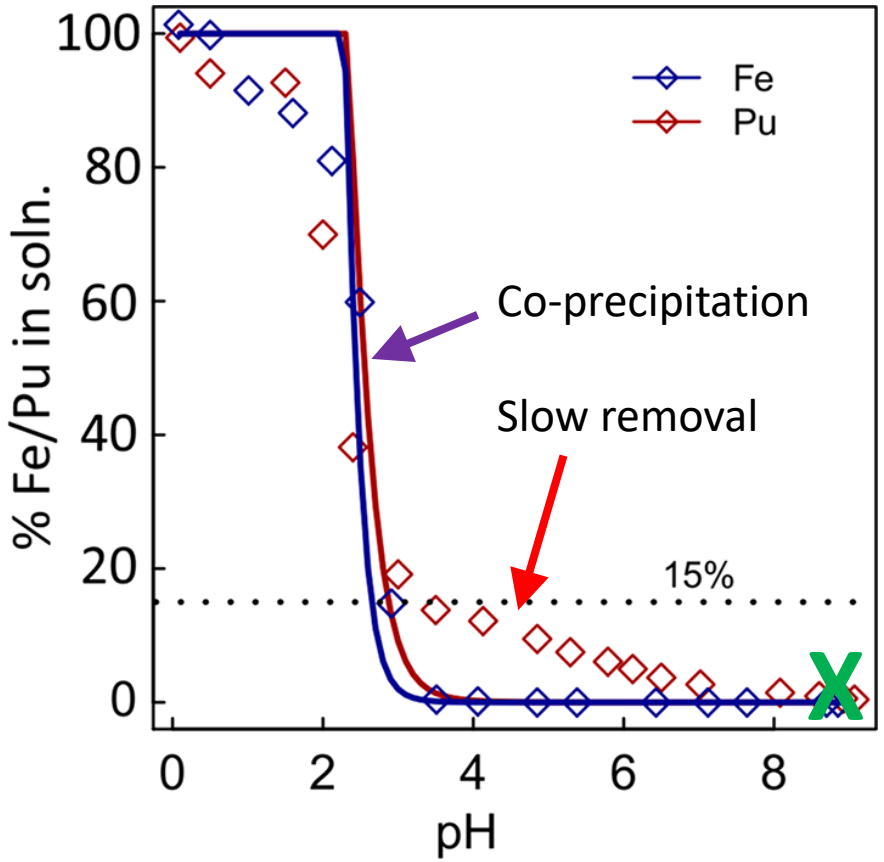


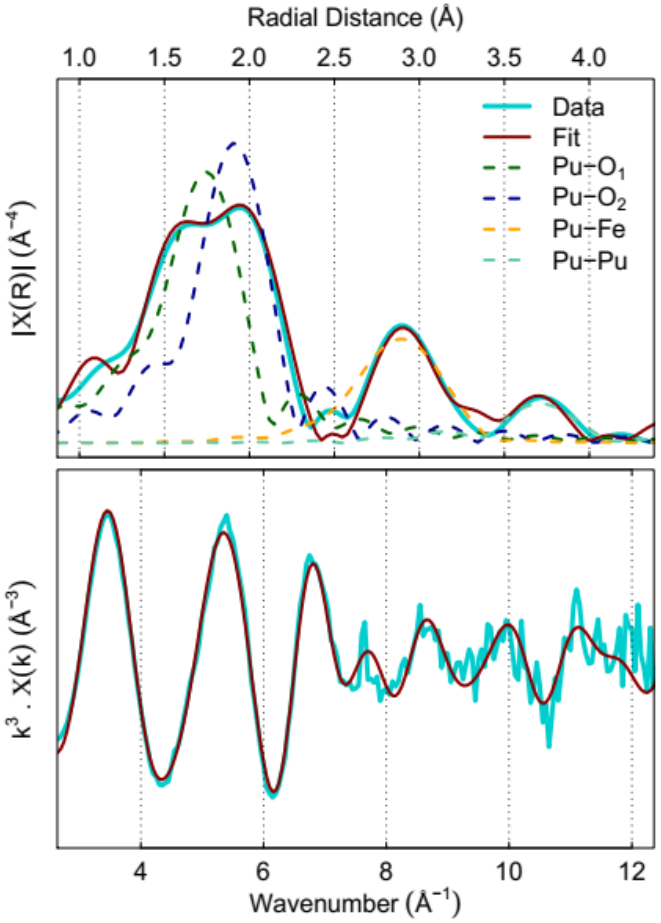
EXAFS – sorption to 10 wt % U;
schoepite precipitation 25 wt % U

Winstanley et al. (2019) Journal of Hazardous Materials 366, 98-104.

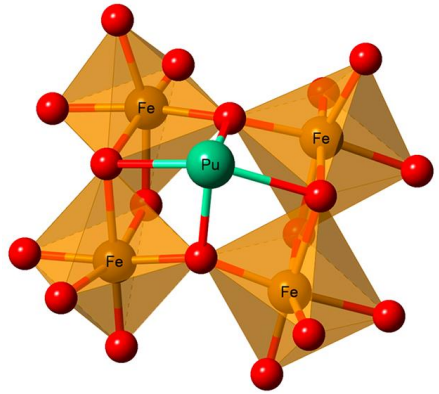
Radionuclide Behaviour in EARP

^{242}Plutonium uptake during ferrihydrite formation – first Pu measurement at DLS





Pu(IV)-sorption complex with modest PuO_2



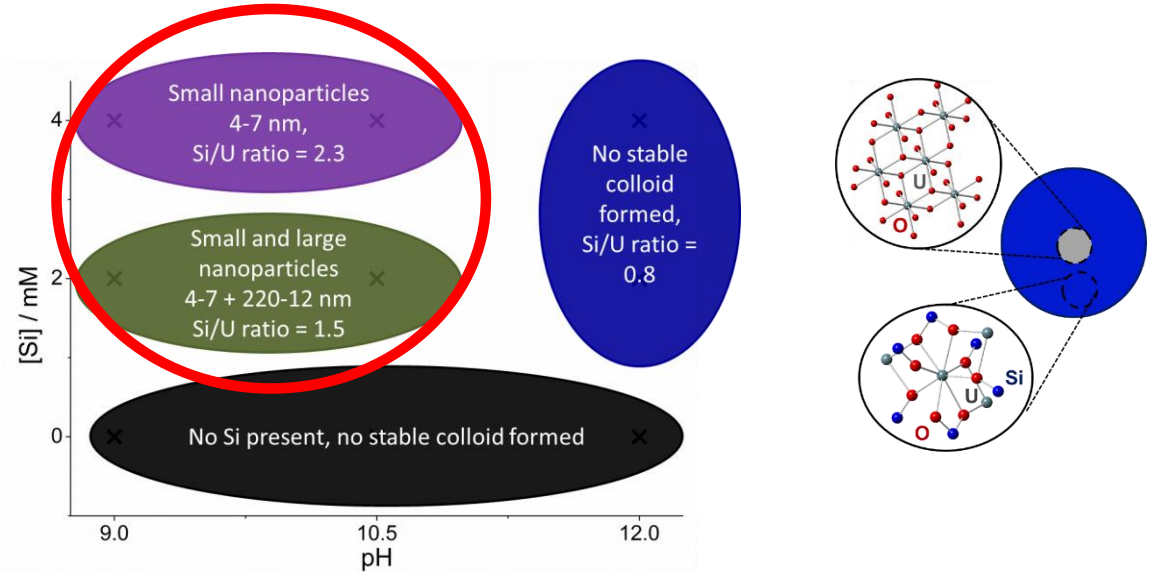
https://pubs.acs.org/doi/10.1021/acsearthspacechem.9b00105

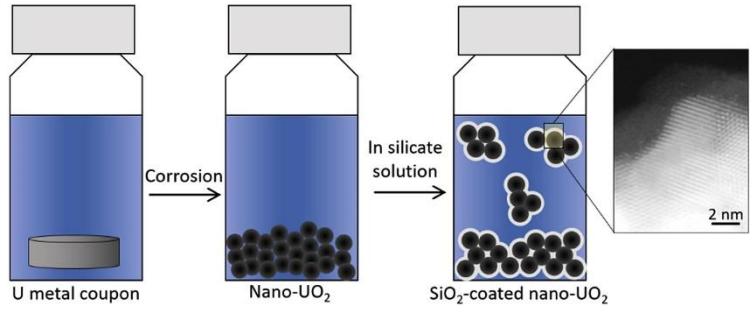
Decommissioning - Sellafield Legacy Ponds and Silos

- Irradiated Magnox fuel metallic uranium → corroded in ponds
- Spent fuel storage ponds: high pH, anaerobic sludge
- Alkaline pond effluent treated in Site Ion Exchange Plant – SIXEP
- U behaviour - colloids

Colloid Behaviour in Ponds - SIXEP

- U(IV)-silicate colloids – stability / structure poorly constrained
- [1] Synthesis of U(IV) colloids and multiple characterization approach - EXAFS, SAXS, PDF, ultrafiltration, DLS, TEM, XRD and zeta potential
- [2] Reaction of uranium corrosion product with Si solutions
- Directly supported case to change settling protocols in SLPS complex → clearer waters, reduced beta challenge





[1] Neill et al., *Environ. Sci. Technol.* 2018, 52, 16, 9118–9127 ; [2] Neill et al., J. Nuclear Materials. 2019, 526, 151751
[3] Foster et al., 2022, Langmuir, doi/10.1021/acs.langmuir.1c03179

NNUF
National Nuclear User Facility

MANCHESTER 1824
The University of Manchester

RADER Facility

- Solid characterisation – XRD, FT-IR, BET
- Aqueous characterisation – Nanosite, zetasizer, UV-vis, ICPMS QQQ, ICPAES
- Mineral characterisation – XRD, eSEM,
- Radiometric characterisation – alpha, gamma and scintillation counting, autoradiography
- Chemostat, cutting and polishing kit, ACs, fume hoods
- Labs: separations, columns, mycology, environmental -mineralogy, -radiochemistry, - geomicrobiology

- People – NNUF EO Tony Stockdale

anthony.stockdale@manchester.ac.uk

Prospect for NNUF AMB

- Growth of user community
  - Extant and new academic users
  - Industry users
- AMB – facilitates access to range of beamlines including new capabilities, DLS model really innovative:
  - Radioactive samples from offsite, increased radiotoxicity, increased knowledge on sample cells etc.
  - Sub-set of time resolved experiments (seconds – years)
  - Remove barriers to new users
- Mechanistic insight proven to be important – communities, regulatory, international safety cases

Funding

diamond
Sellafield Ltd
UKRI Natural Environment Research Council
NATIONAL NUCLEAR LABORATORY
UKRI Science and Technology Facilities Council
UKRI Engineering and Physical Sciences Research Council
MANCHESTER 1824 The University of Manchester
NNUF National Nuclear User Facility