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Synchrotron X-ray analysis in support of radioactive waste disposal and nuclear decommissioning

Prof. Claire Corkhill

Diamond Active Materials Building Launch Seminar, 1st March 2022.

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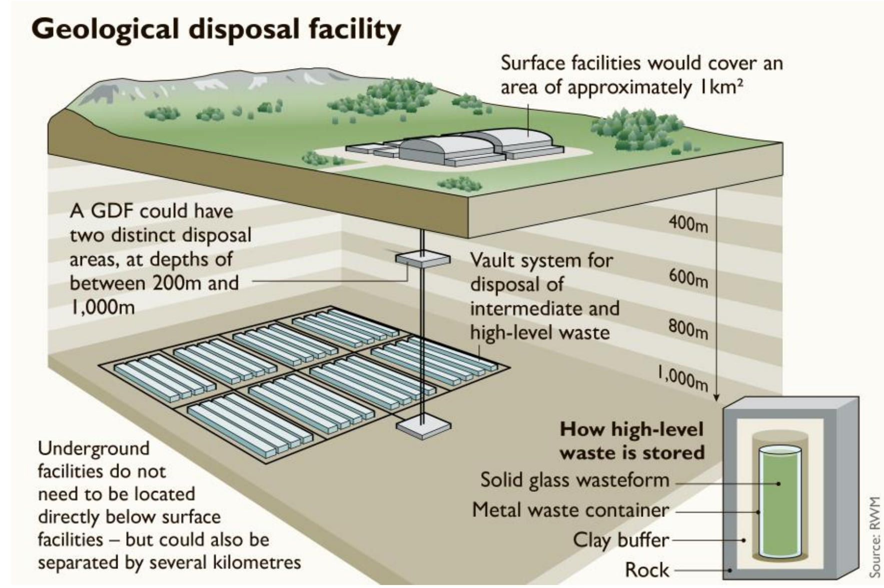
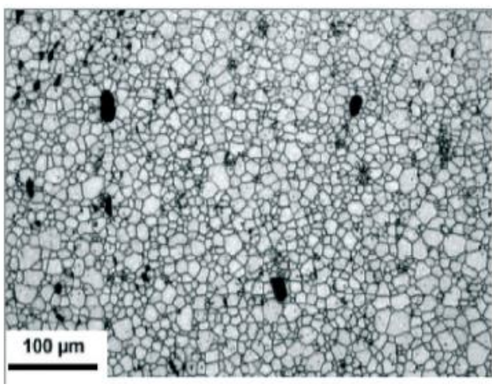
@ISL_Sheffield

Reducing uncertainty in nuclear fuel disposal

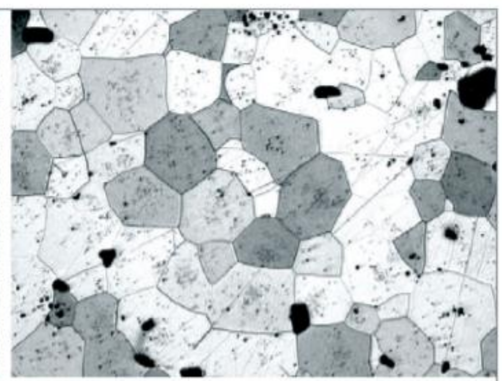
- Release of radionuclides to GDF environment controlled by rate of fuel dissolution by groundwater
- UO_2 corrosion rates well understood



Y. Guerin, CEA-DEN Monographie, Nuclear fuels, **2009,** p47.

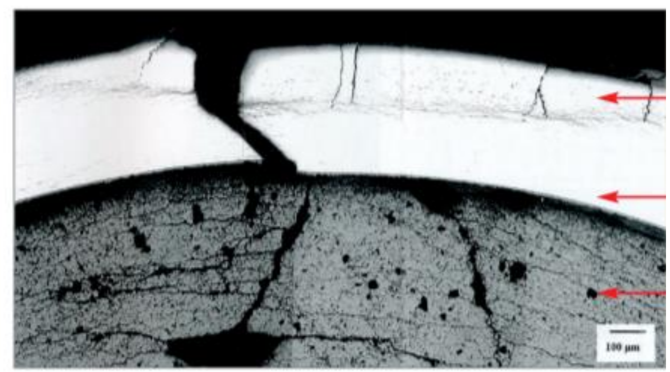


UO_2 grain size ≈ 10 µm



Cr-doped UO_2 Grain size ≈ 50 µm

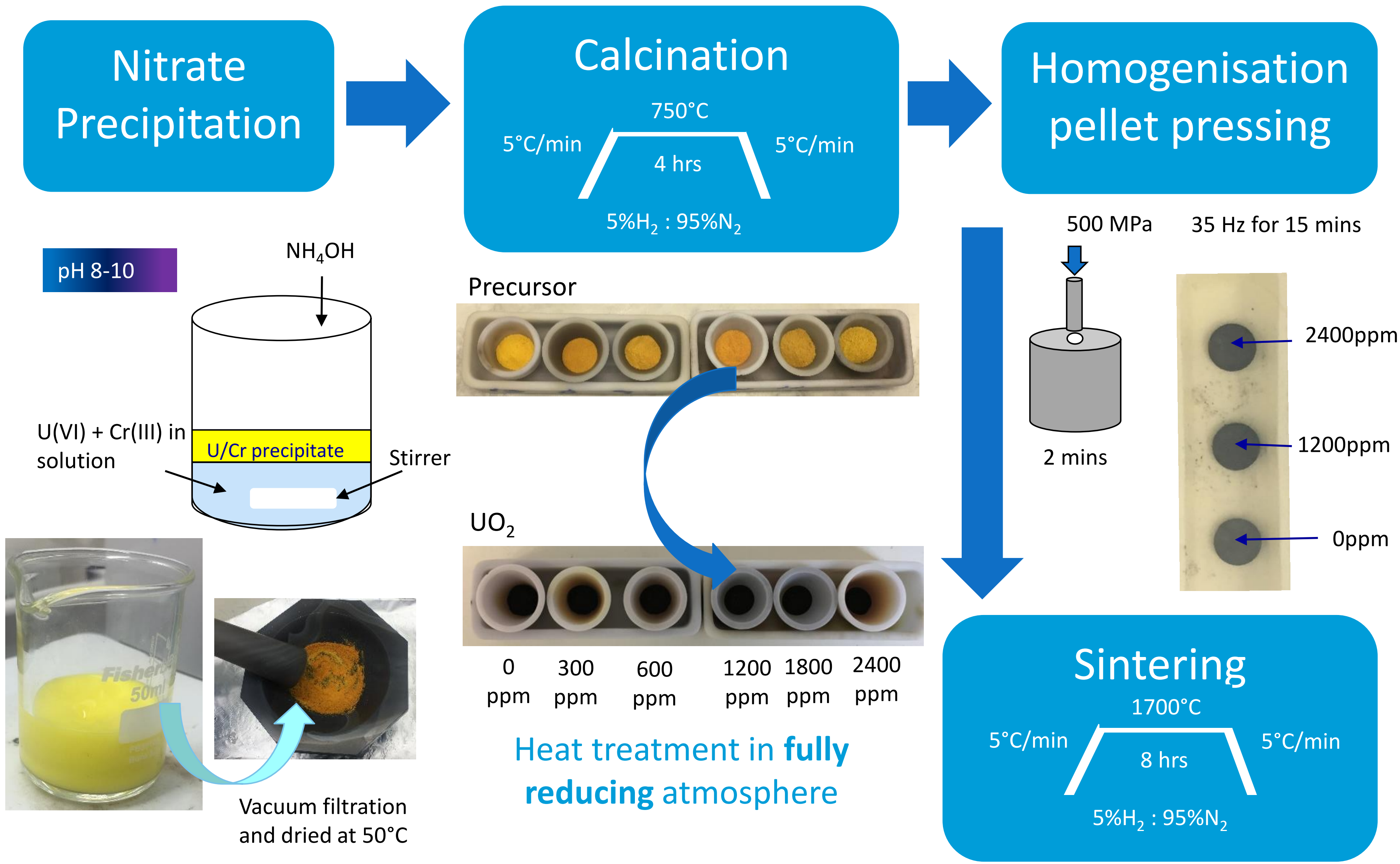
G. Ducros, CEA-DEN Monographie, Nuclear fuels, **2009,** p65.



Cladding failure due to fission gas build-up

Chemistry of Cr-doped UO_2 not well understood. Will the addition of Cr influence the long-term durability in a geological disposal facility?

Cr-doped UO_2



Smith et al. Nature Materials, IN REVIEW

Cr K-edge X-ray absorption spectroscopy at I20

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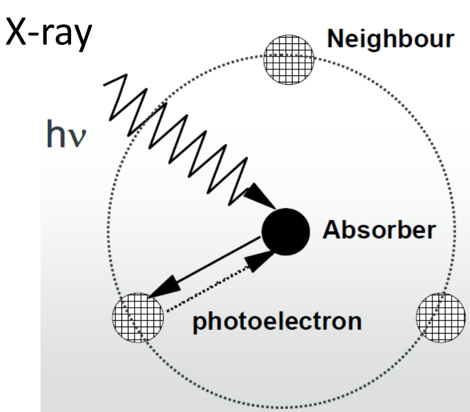
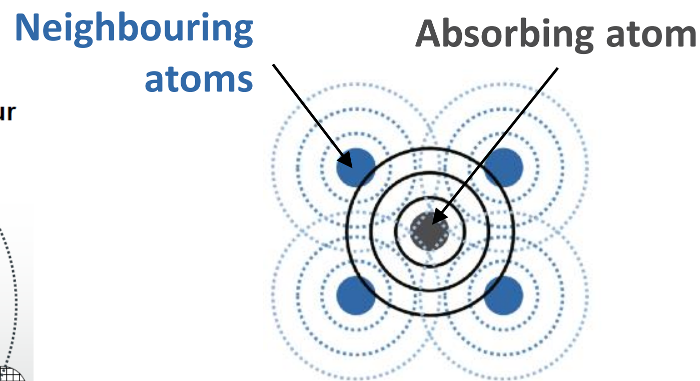
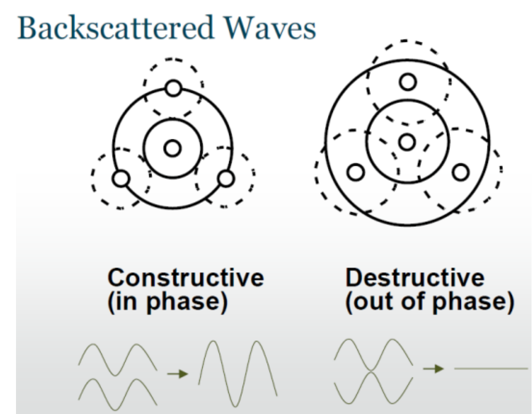
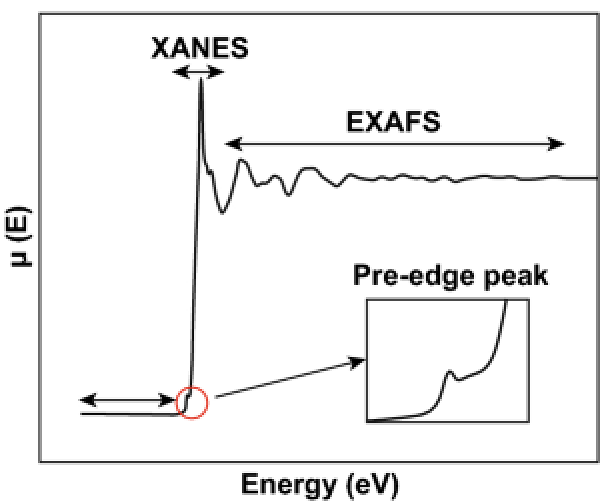




Interference pattern of outgoing and reflected photoelectron waves





X-ray Absorption Near-Edge Structure → sensitive to oxidation state and electronic structure

Extended X-ray Absorption Fine Structure → contains information about the local atomic structure such as bond distance and coordination number.

Cr K-edge XAS at I20

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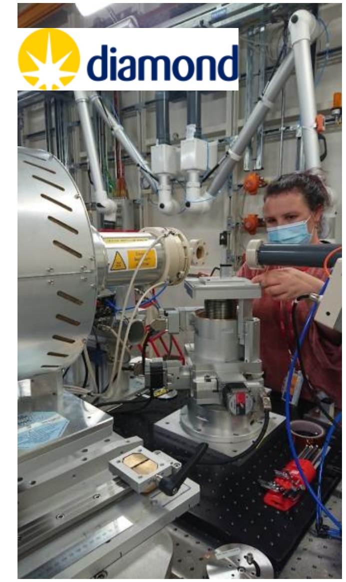
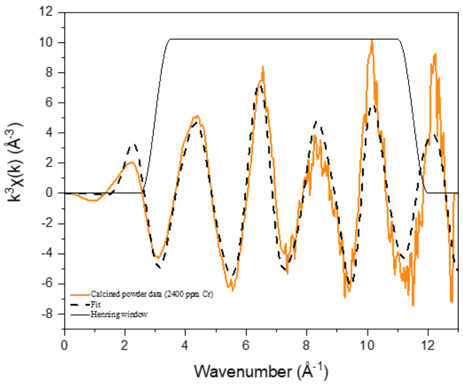
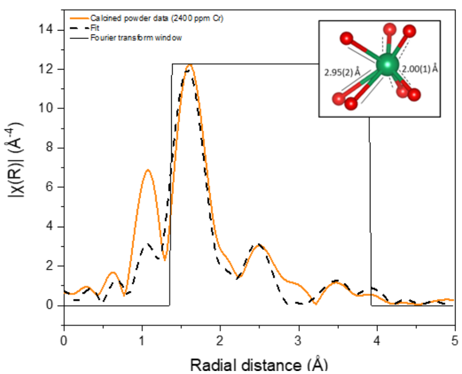
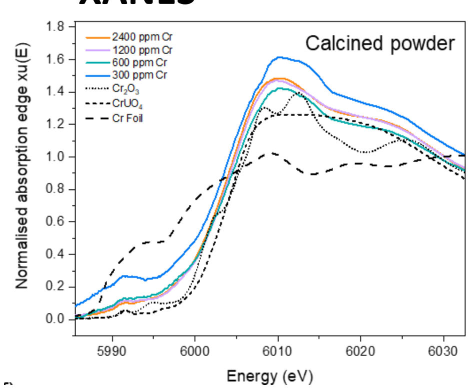


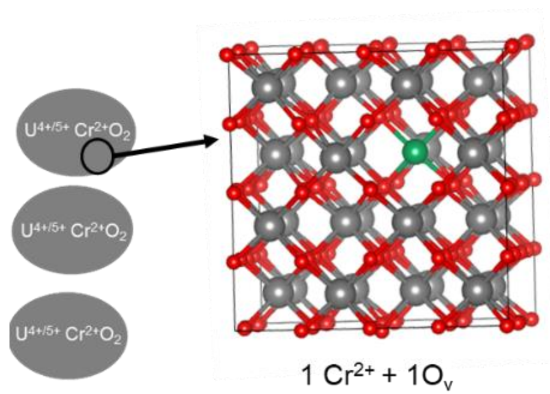
EXAFS



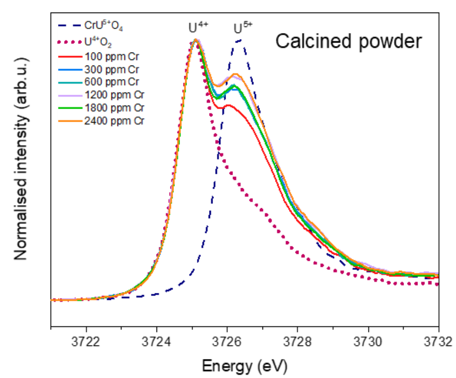


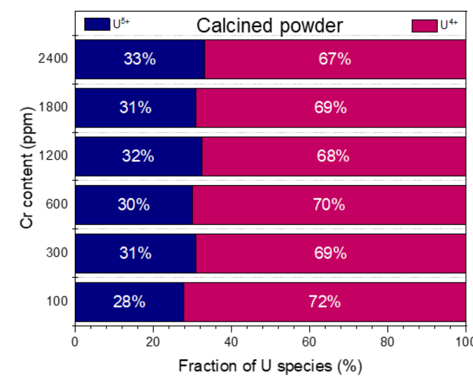
XANES





HERFD-XANES (ESRF)





Smith et al. Nature Materials, IN REVIEW

Cr^{2+} dopant influences U dissolution rate in simple groundwater

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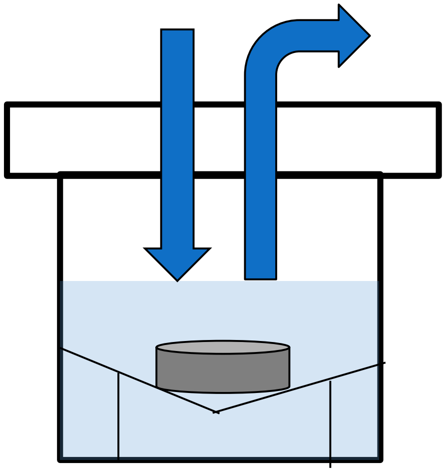
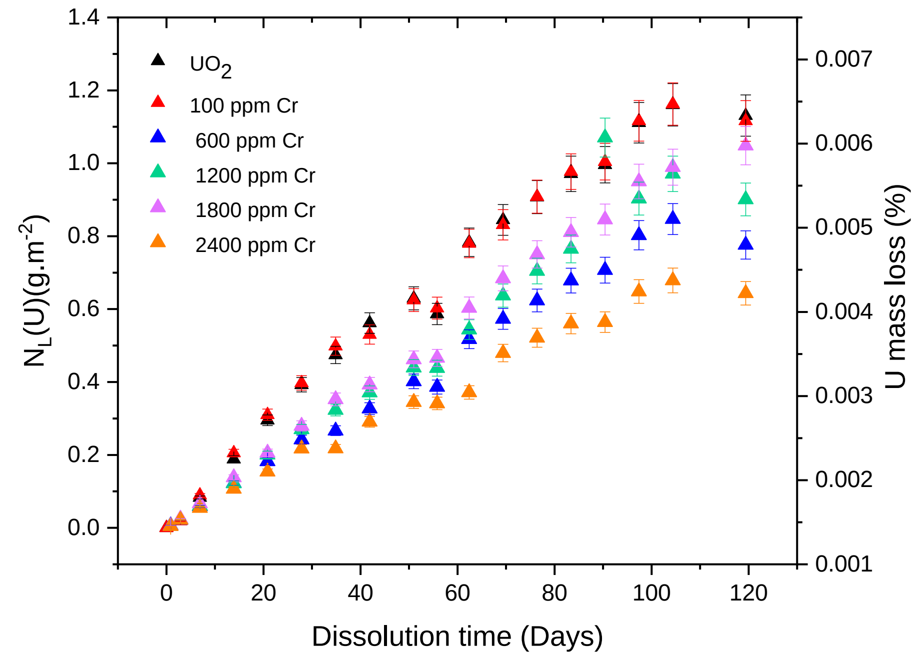
HENRY ROYCE INSTITUTE NNUF National Nuclear User Facility PLEIADES



Remove and replace aliquot of solution for analysis via ICP-MS



ICP-MS measures concentration of U and Cr in solution

$$C(i) = \frac{m(i)}{V}$$

Mass element in solution

Volume of solution

Normalised mass loss

$$N_{L(i)} = \frac{M(i)}{f(i) \times S}$$

Mass fraction of element

Surface area

Hypothesis: Galvanic coupling of Cr with $U^{4+}/U^{5+}/U^{6+}$ reduces rate of U dissolution.

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Wouldn't it be awesome if there were somewhere we could run long-term corrosion experiments at Diamond, to be able to monitor chemical degradation, *in-situ*?

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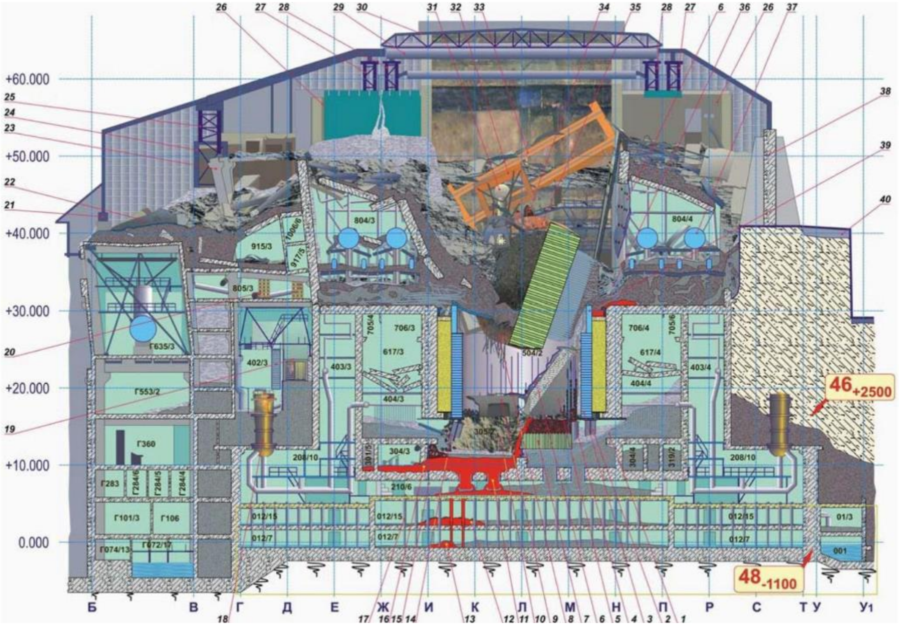
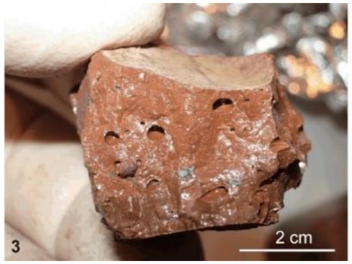
Nuclear fuel debris

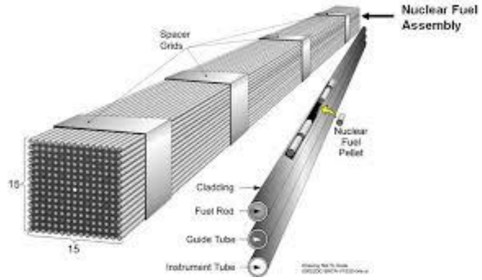
Chernobyl Reactor 4 (1986)



Brown Lava



Black Lava



$U_{1-x}Zr_xO_2$ solid solution

$U_{1-x}Zr_xO_2$ + Concrete + Stainless steel

Decommissioning requires:

- Understanding of fuel chemistry
- Knowledge of mechanical properties
- Evaluation of corrosion mechanisms and generation of α-active dust

LFCM (Lava-like Fuel Containing Material)
MCCI (Molten Core-Concrete Interaction)

Pictures courtesy of Boris Burakov.

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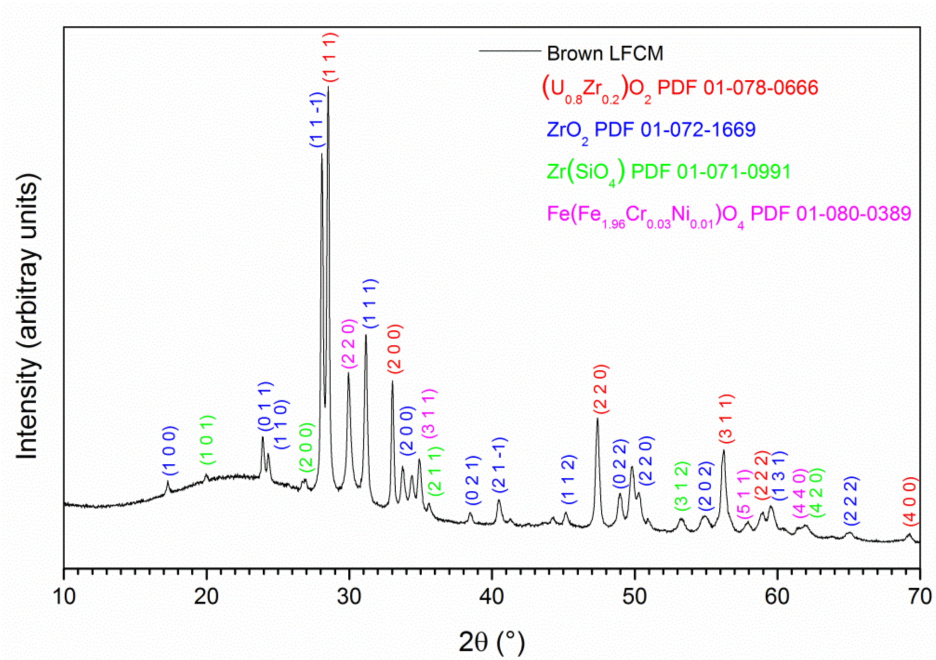
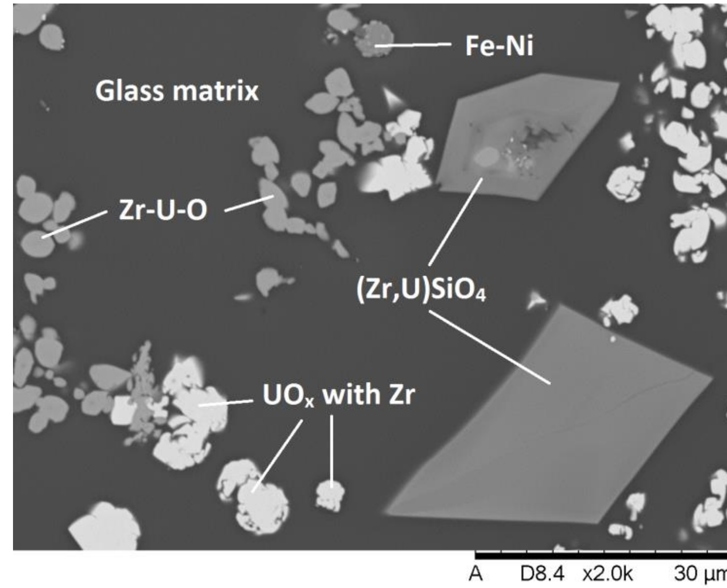
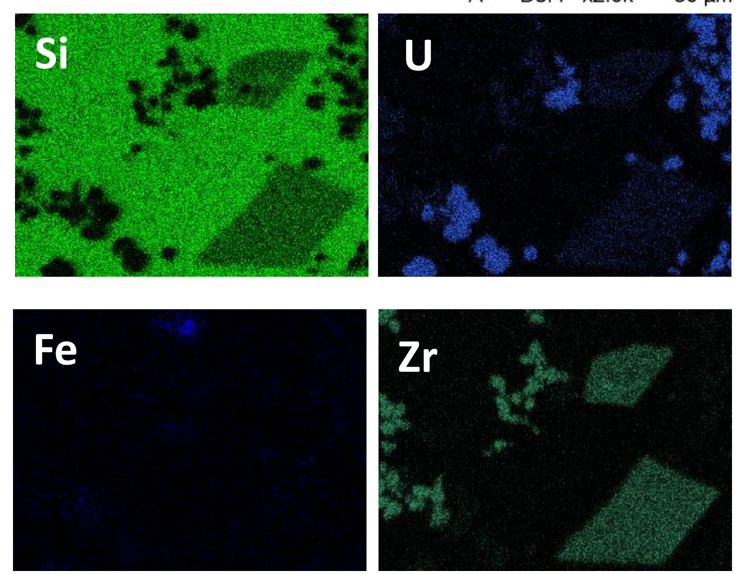
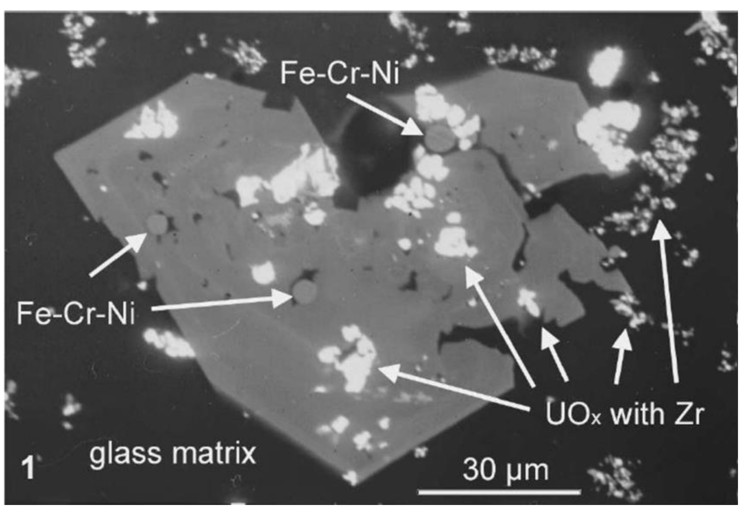
Simulant Chernobyl fuel debris









“Real” LFCM by Boris Burakov

Anderson et al. Radiochimica Acta, 60, 149 (1993)

Chernobyl: Barlow et al. npj Mater. Degrad. (2019); Ding et al. JMCA (2021), Fukushima: Ding et al. npj Materials Degradation (2022)

Nuclear fuel debris corrosion

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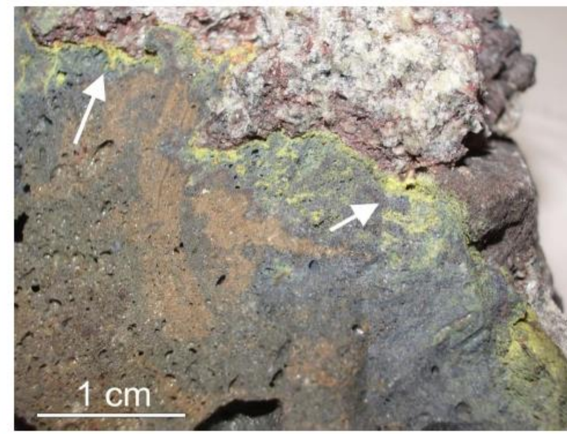
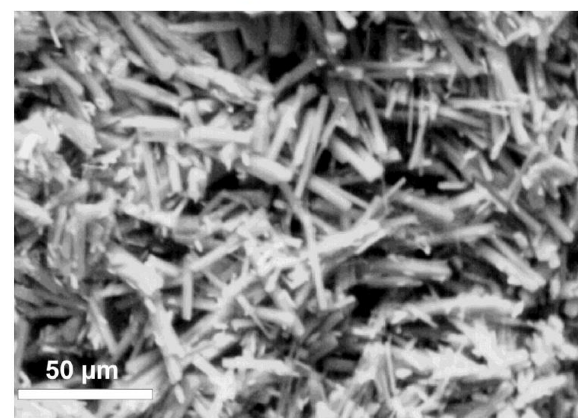


Uranium corrosion in fuel debris at Chernobyl resulted in generation of hazardous α-containing dust → respirable hazard during decommissioning

- $Na_3H(CO_3)_2 \cdot 2H_2O$
- $UO_3 \cdot 2H_2O$
- UO_2CO_3
- $Na_4(UO_2)(CO_3)_3$
- $UO_4 \cdot 4H_2O$ (studtite)



Pictures courtesy of Boris Burakov.

Unique long duration facility

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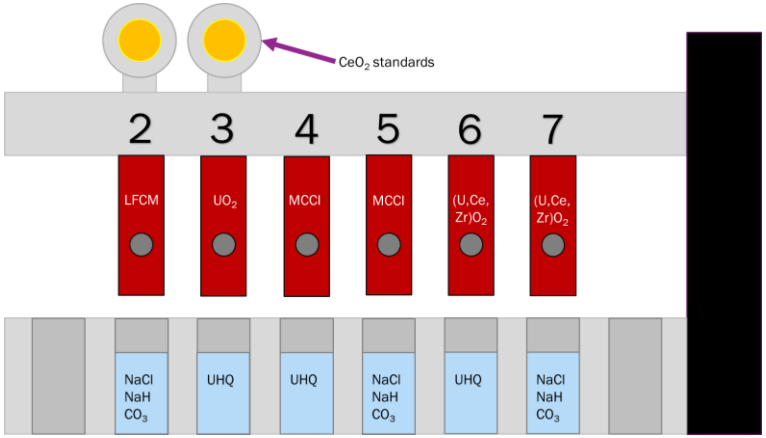
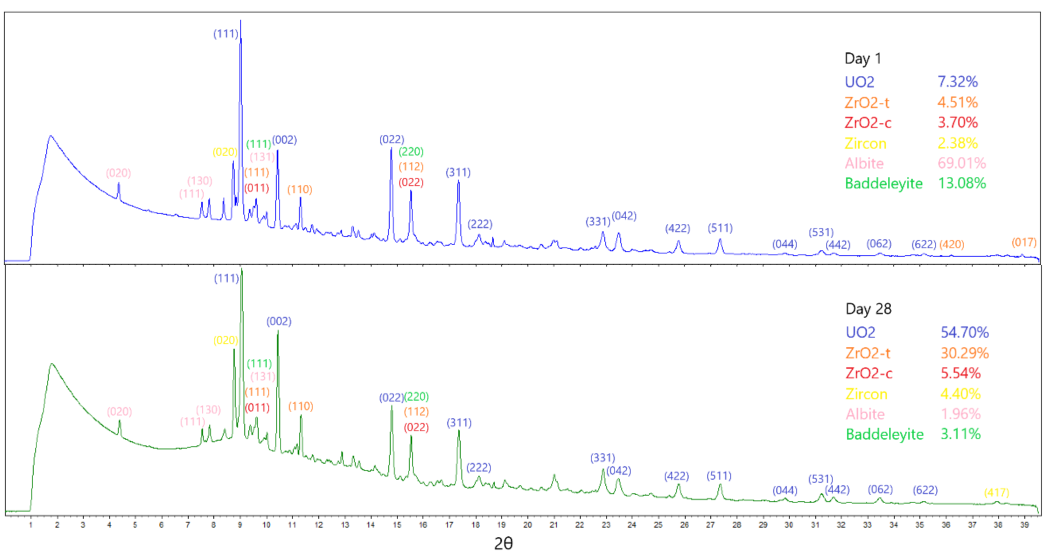
Beamline I11-LDE X-ray Diffraction



Weekly diffraction patterns acquired from UO_2 SIMfuel and Chernobyl / Fukushima fuel debris, during *in-situ* corrosion



Opportunities for AMB

Access to laboratory space for running mid to long-term degradation experiments, with samples periodically monitored by synchrotron techniques (subject to successful beamtime application)

Ideal if degradation of materials could be in air, aqueous environments, variable temperature (ovens up to 90°C), or controlled atmosphere, etc.

Shorter-term degradation also of interest, e.g. oxidation of advanced technology fuels in air. Access to AMB essential for preparation of beamline samples over a fixed period of time.

Thank you for your attention!

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