Transitioning of Spent AGR Fuel from Wet to Dry Storage

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Introduction

- Large quantities of nuclear fuel has accumulated in storage ponds.
- Fuel can not be stored in ponds indefinitely.
- In many countries dry storage of spent nuclear fuel (SNF) is used as an interim measure.
- Key requirements for dry storage are criticality prevention, integrity maintenance and retrievability.
- All of these are affected by corrosion and hence the interaction with water.

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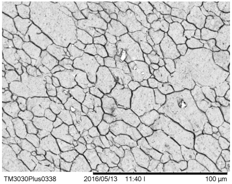
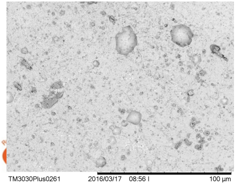
Work to Date

Corrosion Behaviour

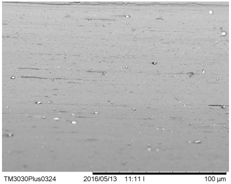
- Differences in corrosion behaviour of inner and outer cladding surface.

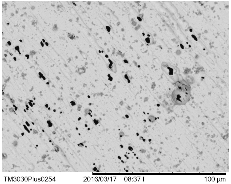
Inner Surface



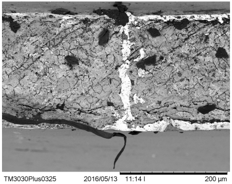


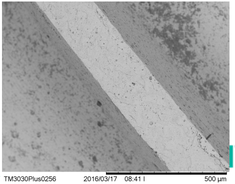
Outer Surface





Rib



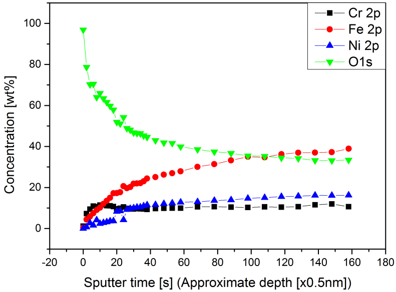


Work to Date

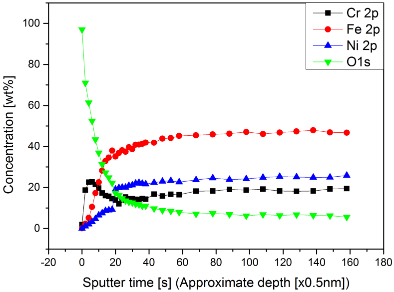
XPS Depth Profiling

- Outer surface appears to have lower oxygen content and thinner oxide layer.

Inner

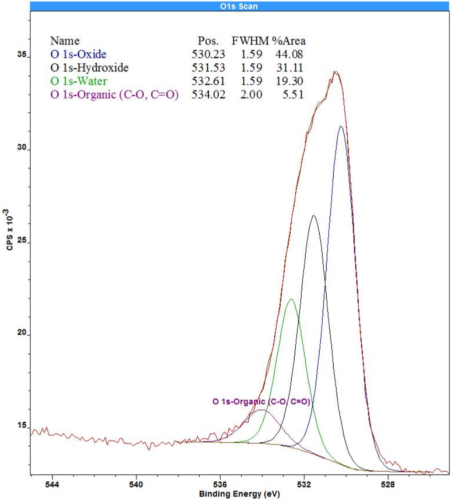


Outer

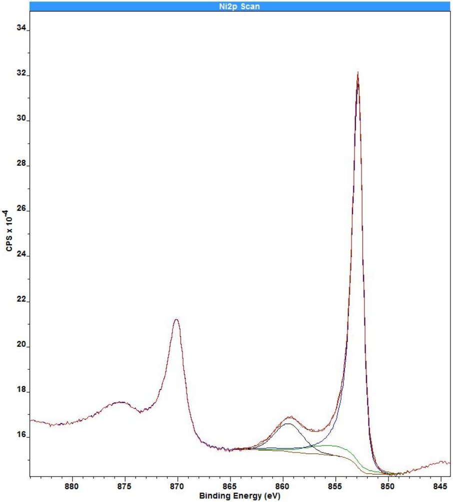


Work to Date

Oxygen

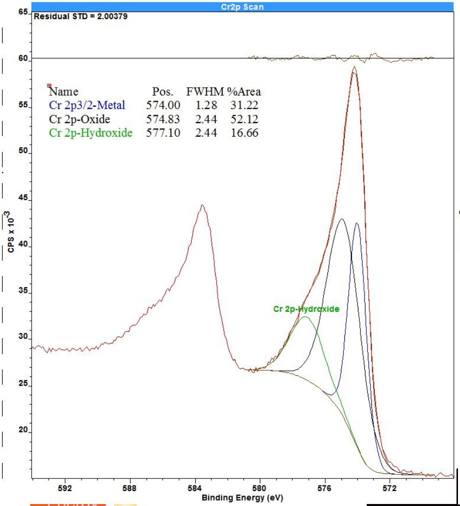


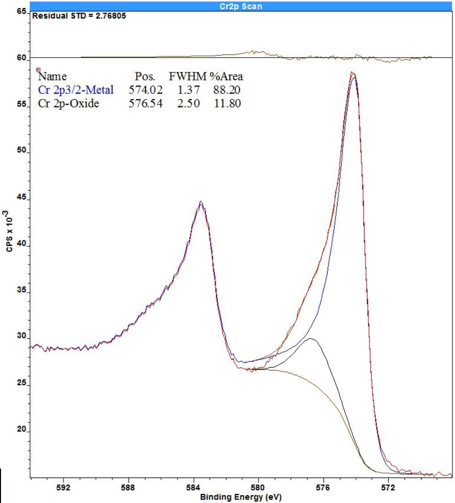
Nickel



Work to Date

Chromium. Expected peaks 574.3, 576 and 577.3 eV for metal, oxide and OH respectively.





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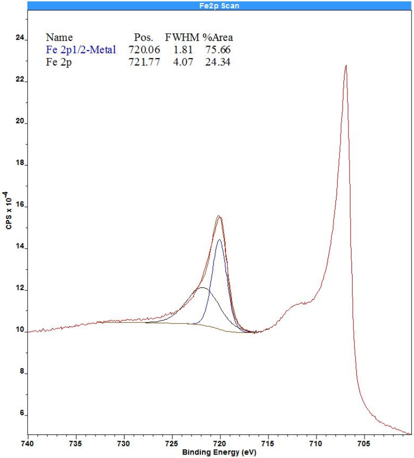
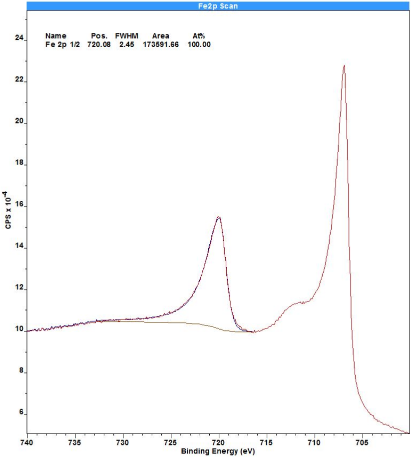
Work to Date

Iron. 3/2 peak overlaps with Ni LMM Auger.



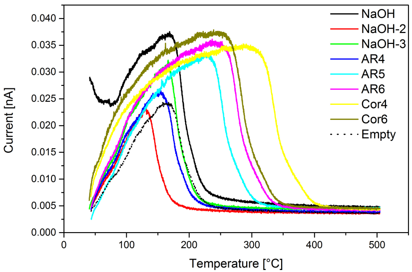


Work to Date

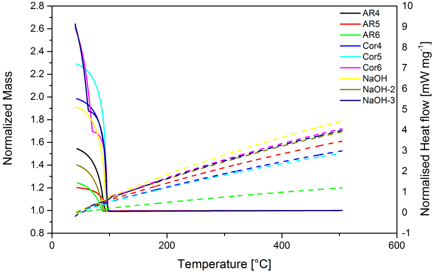
Adsorbed Water

- TGA/MS Shows insignificant adsorbed water.

MS

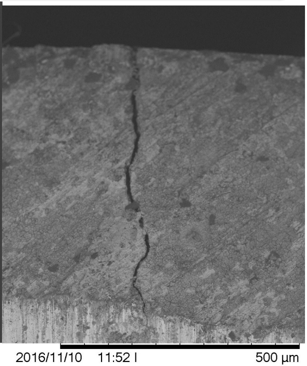


TGA/DSC



Cracked Samples

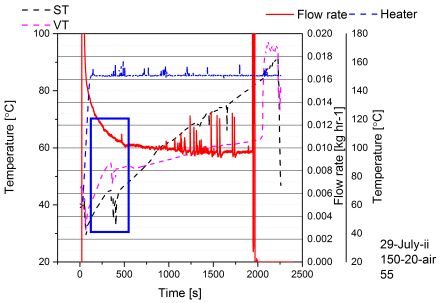
- Steel treated to form cracks.
- No success with 20/25 but have cracked 304.

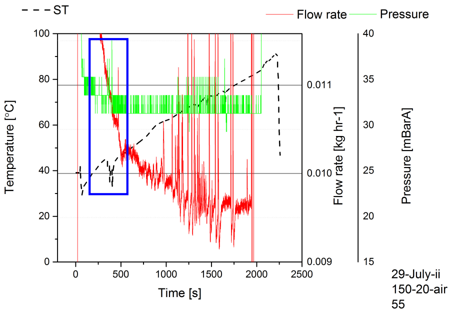


Drying Rig and Test Piece

Drying Rig Data

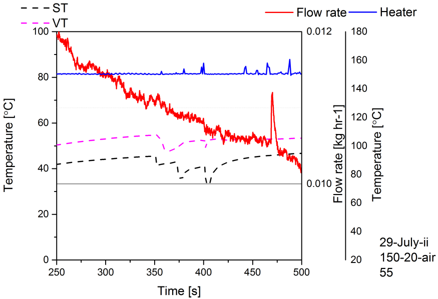
Data output is something like this.

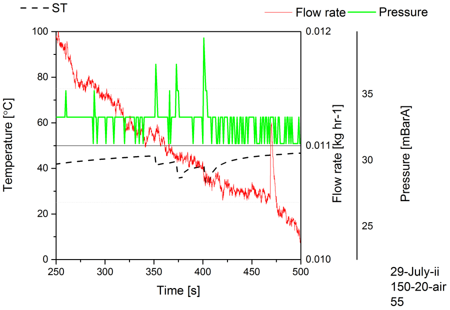




Drying Rig Data

Zooming in on highlighted areas.



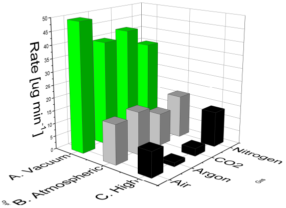


Work to Date

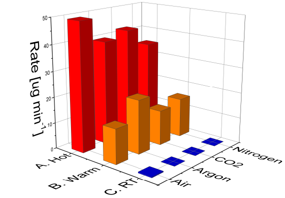
36 Different Conditions Used

- Three Temperatures; RT, 60°C (wam) and 150°C (hot).
- Three Pressures; 20 mBarA, atmospheric pressure, high pressure (10 BarG).
- Four different gases; air, N_2, Ar and CO_2.

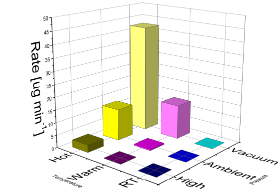
Hot drying



Vacuum Drying



Single gas-CO_2



Summary

Sample Preparation

- Adsorbed water is negligible.
- Still working on XPS and SCC.

Drying Tests

- Initial work has shown hot vacuum drying to be most effective.
- Further tests ongoing.

Grain Boundary Damage Mechanisms in strained AGR under Irradiation

C. Barcellini*[1,2], S. Dumbill[3], S. Pimbott[1,4], and E. Jimenez-Melero[1,2]

[1]DCF, UoM, [2]School of Materials, UoM, [3]NNL, UoM, [4]School of Chemistry, UoM
(*Correspondence:chiara.barcellini@postgrad.manchester.ac.uk)

DISTINCTIVE Theme 1 Meeting: AGR, Magnox and Exotic Spent Fuels
The Rheged Centre, Redhills, Penrith, 15th November 2016

Contents

Contents

Interim Storage of AGR Spent Fuel

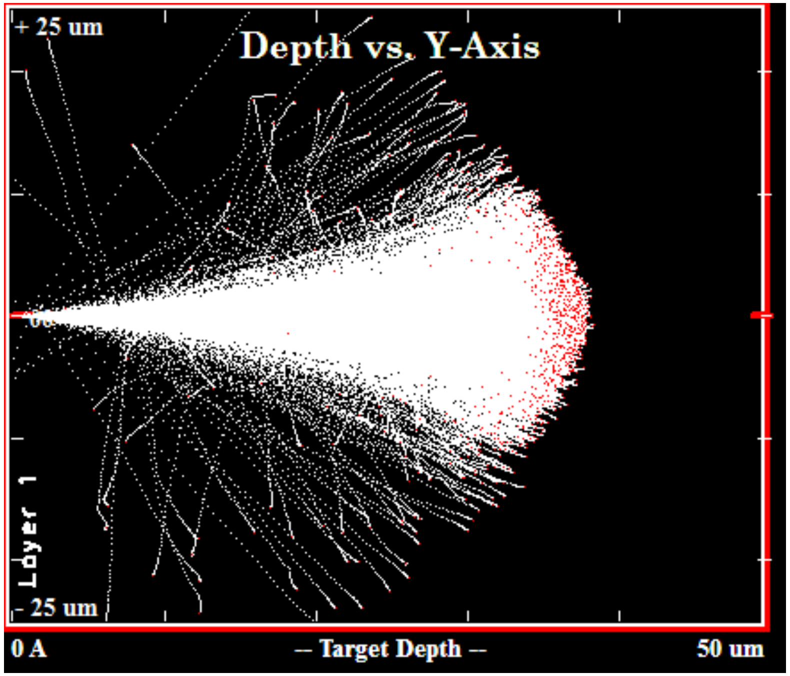
- Wet storage of AGR fuel might last several years
    - challenge for the corrosion properties of the cladding material
- Through-wall failures during wet storage are ascribed to inter-granular corrosion (IGA)
- Lack of recent a about irradiated AGR cladding material
- Difficulties in studying neutron-irradiated materials

Sensitisation with charge particles

Sensitised specimens can be produced using intense beams of protons or heavy ions

ADVANTAGES:
- user-adjustable radiation
- similar microstructure
- reduced induced radioactivity

PhD main Objectives:
- access irradiation conditions
- study microstructures produced during irradiation



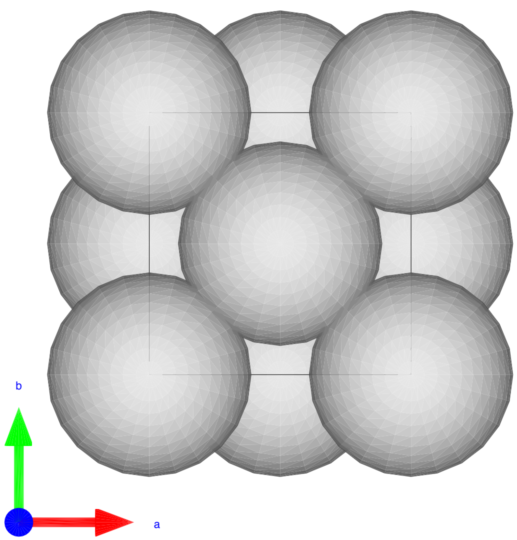
Contents

AGR cladding material

20Cr25Ni
Nb-stabilised
s.s.

Austenitic Stainless Steel
Fe-Ni-Cr alloy

- high corrosion resistance
- no ductile-brittle transition
- high creep resistance



Austenitic matrix has an FCC crystal structure

Specimen Preparation

20Cr25Ni Nb-stabilised s.s.

- cold work
- 40min at 930°C

Annealed specimen

- irradiation

Irradiated specimen

Annealed specimen:

- austenitic matrix
  - composition -> SEM
  - g.b -> SEM & EBSD & TEM
  - texture -> EBSD
- second phases
  - composition -> SEM & TEM
  - texture -> EBSD

Irradiated specimen:

- austenitic matrix
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  - g.b -> SEM & EBSD & TEM
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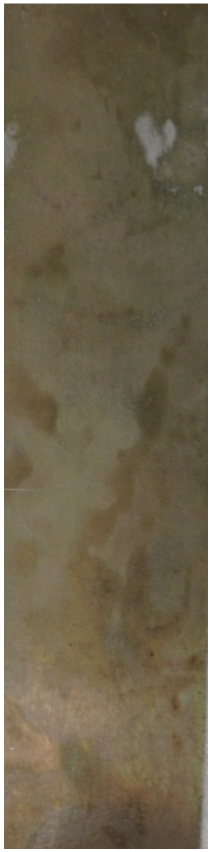
- irradiation

Irradiated specimen

- austenitic matrix
  - composition -> SEM
  - g.b -> SEM & EBSD & TEM
  - texture -> EBSD
- second phases
  - composition -> SEM & TEM
  - texture -> EBSD

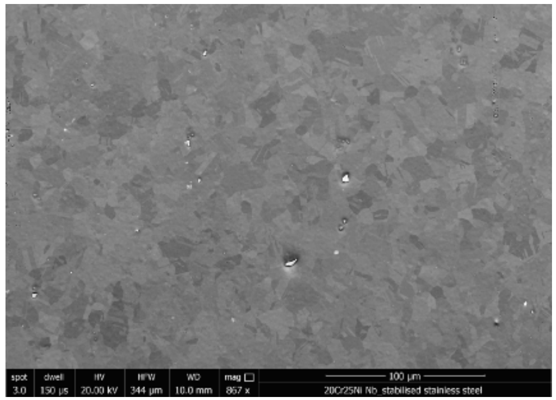
Materials as received

BATCH 1
930°C for 40min

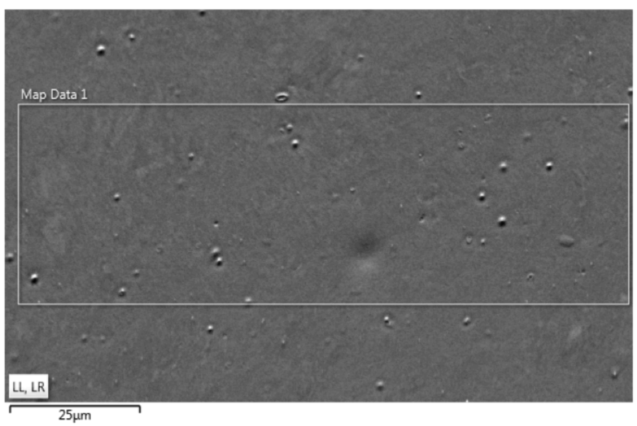


94.17 mm

26.27 mm



BATCH 2

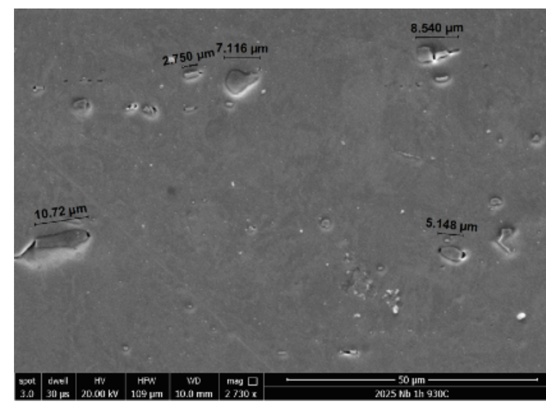


550 mm

16 mm

AGR TUBE

- cut
- outer ribs removed (0.5mm thick)
- split
- rolled until flat



- heat treat 930°C for 40min

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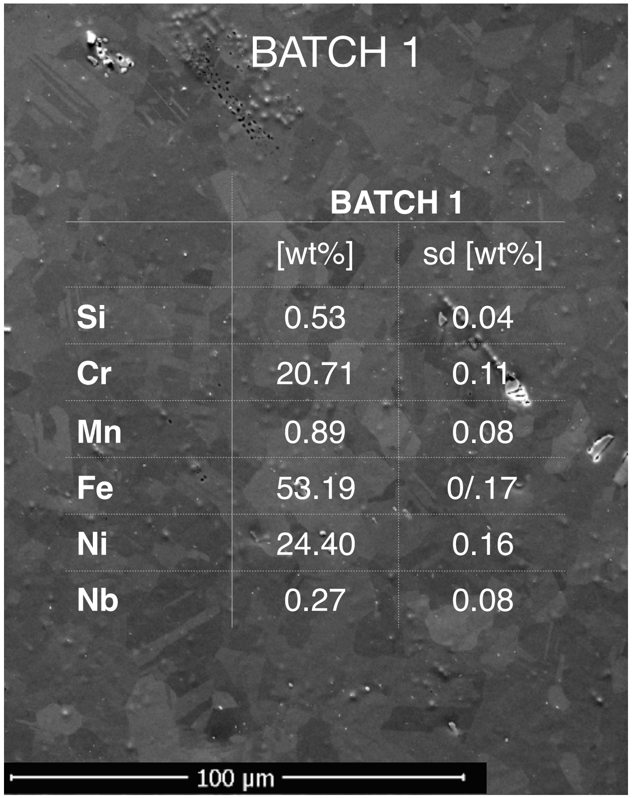
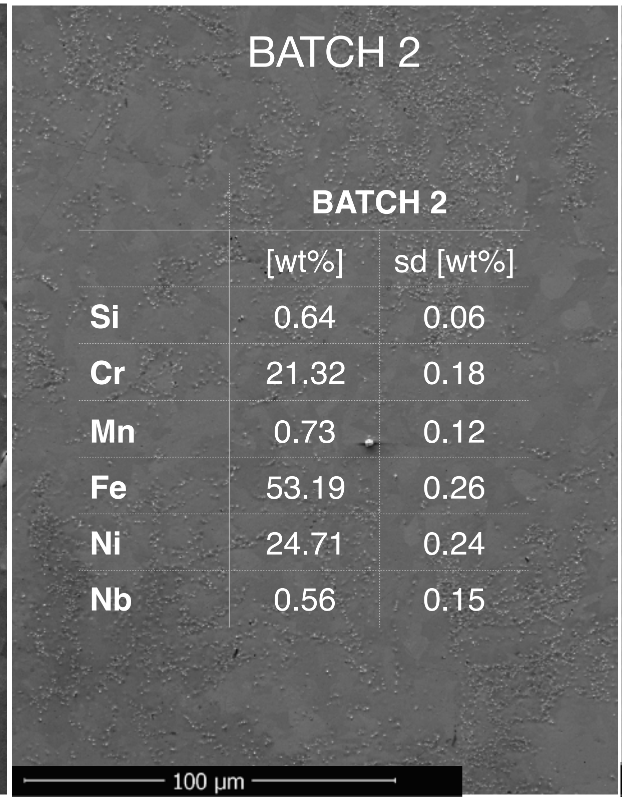
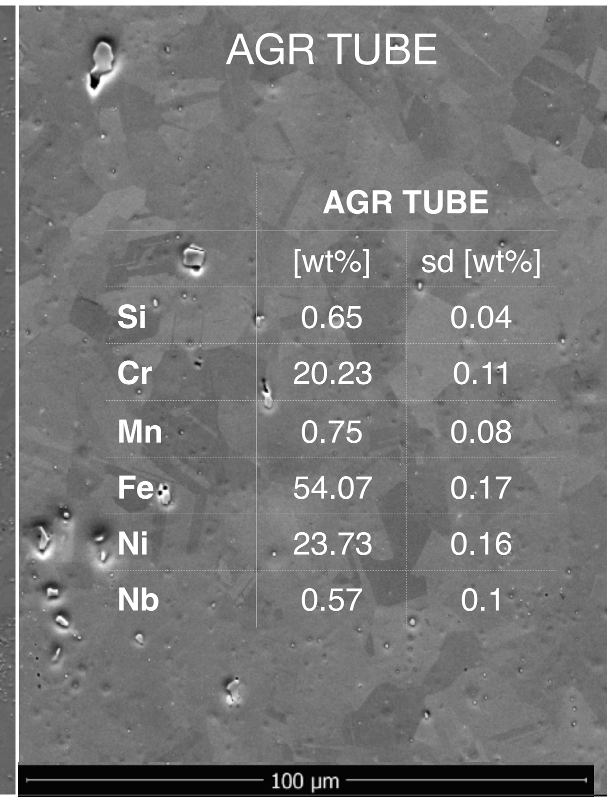
Annealed 20Cr15N Nb-stabilised s.s.



	BATCH 1	
	[wt%]	sd [wt%]
Si	0.53	0.04
Cr	20.71	0.11
Mn	0.89	0.08
Fe	53.19	0/.17
Ni	24.40	0.16
Nb	0.27	0.08

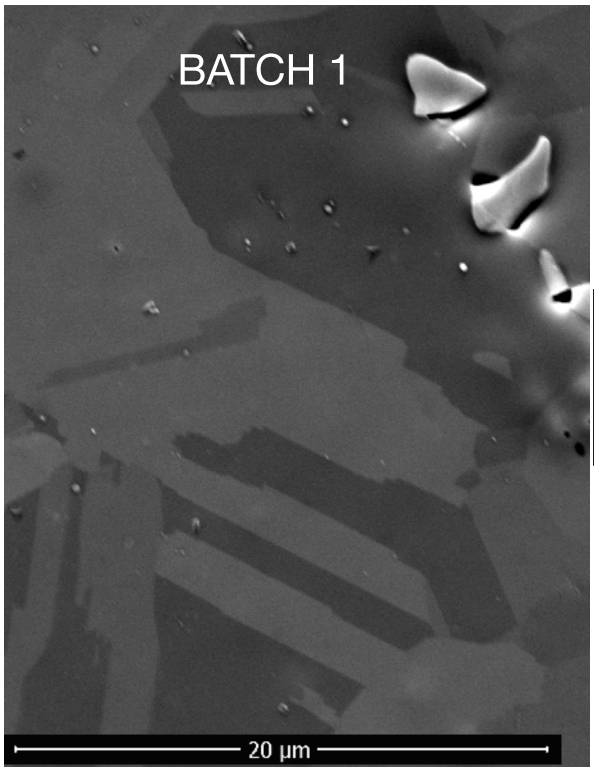


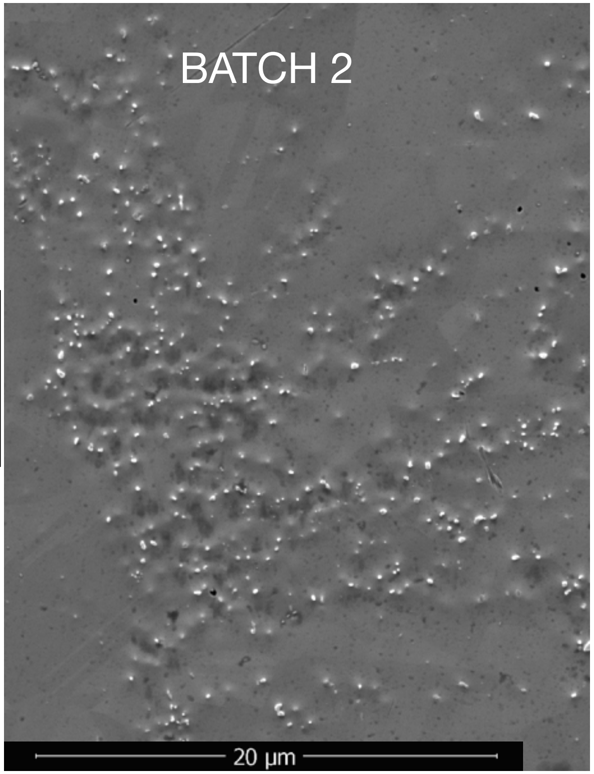
	BATCH 2	
	[wt%]	sd [wt%]
Si	0.64	0.06
Cr	21.32	0.18
Mn	0.73	0.12
Fe	53.19	0.26
Ni	24.71	0.24
Nb	0.56	0.15

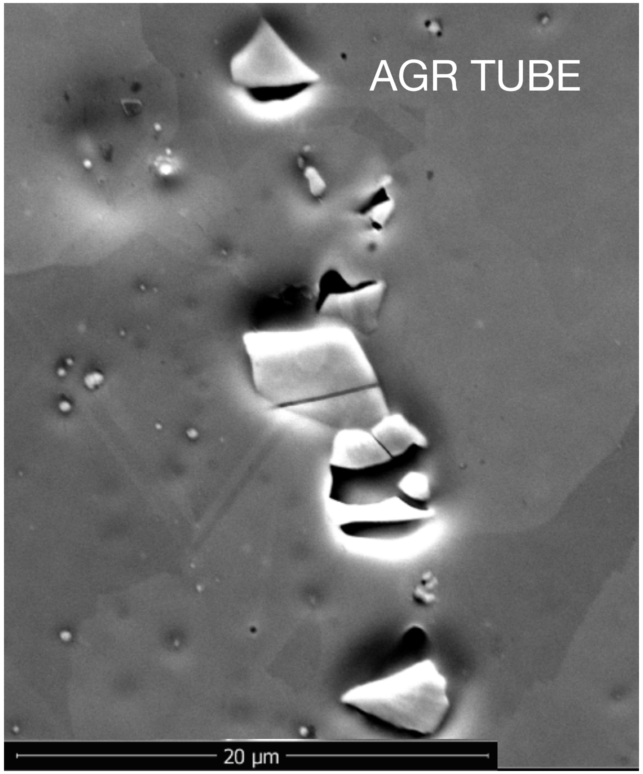


	AGR TUBE	
	[wt%]	sd [wt%]
Si	0.65	0.04
Cr	20.23	0.11
Mn	0.75	0.08
Fe	54.07	0.17
Ni	23.73	0.16
Nb	0.57	0.1

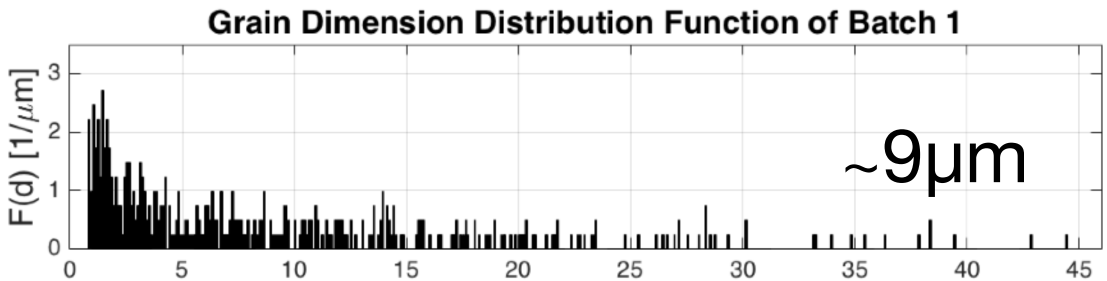
Annealed 20Cr15N Nb-stabilised s.s.

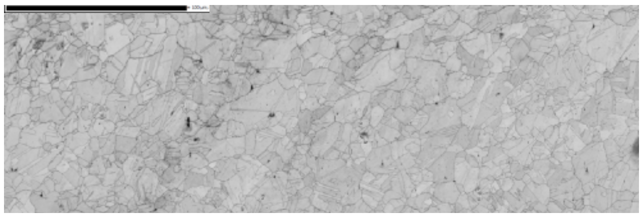


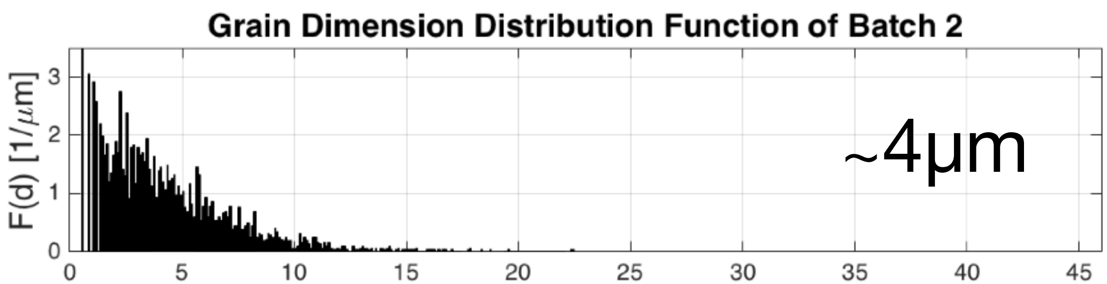


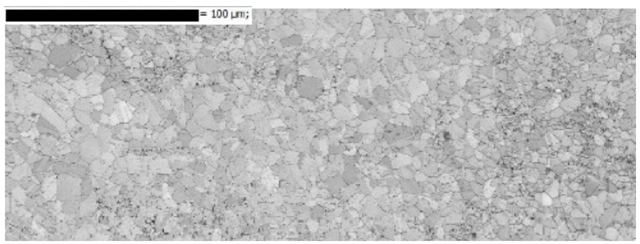


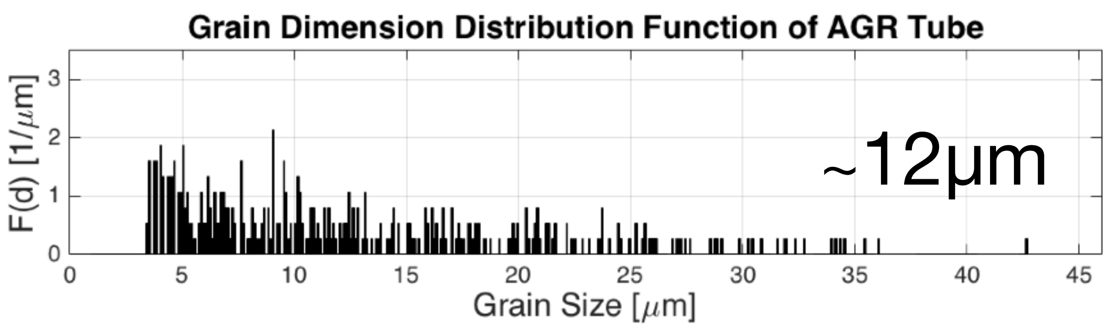
Recrystallisation

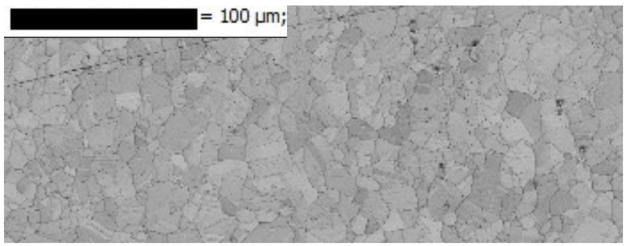




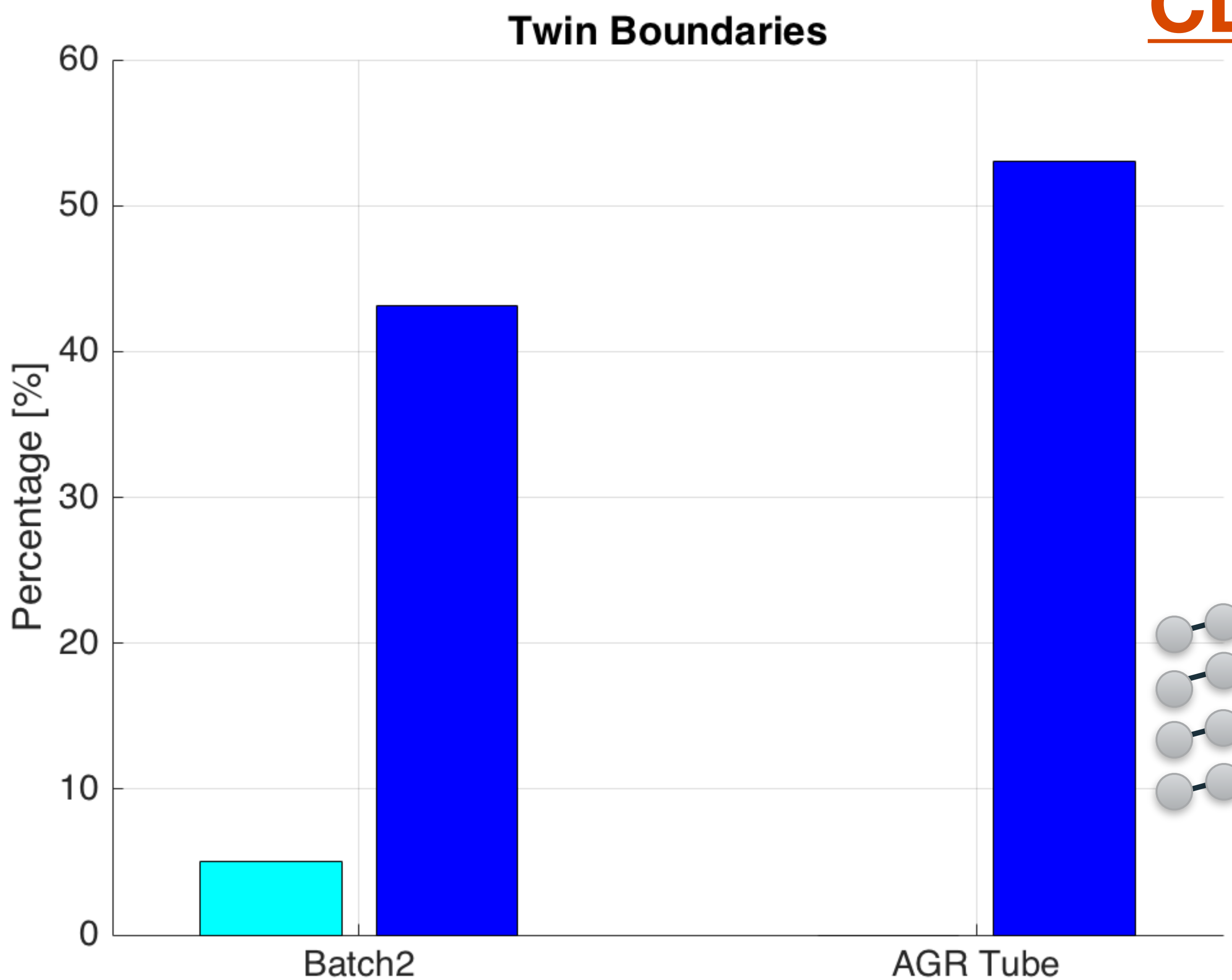


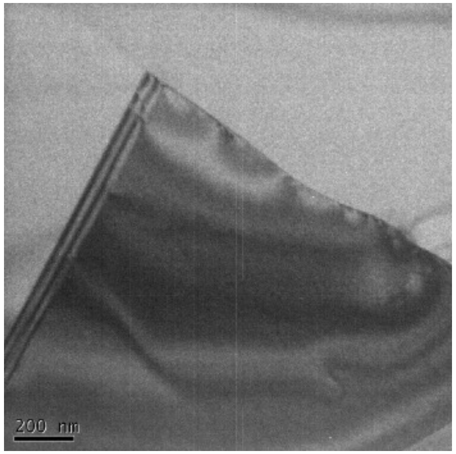


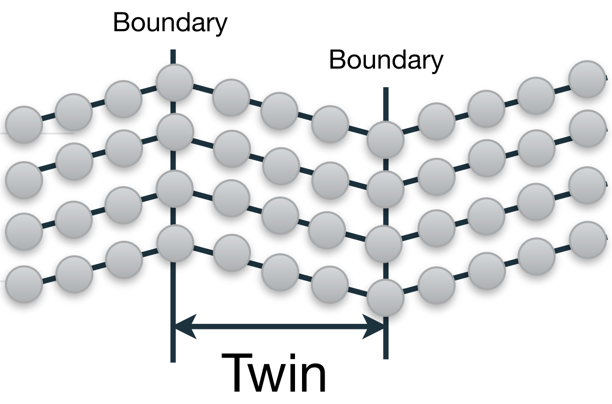




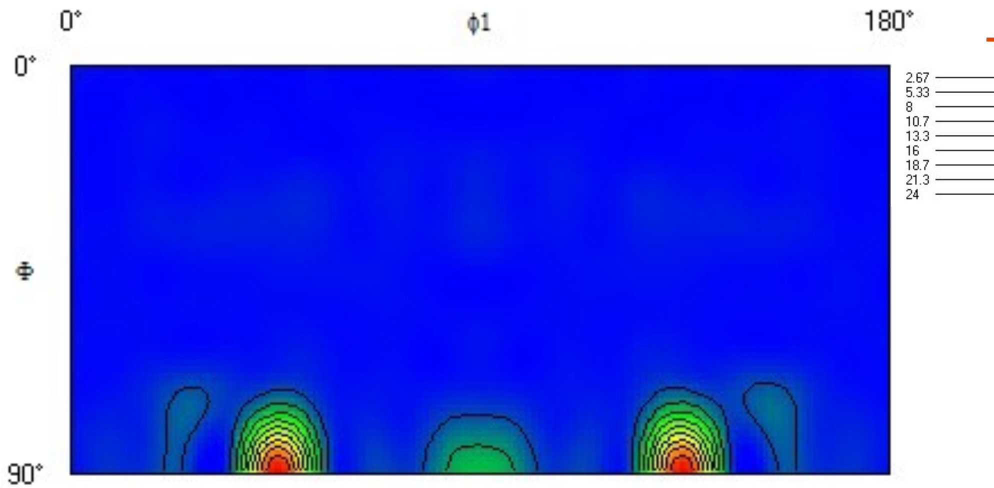
CLS Boundaries







Texture Development

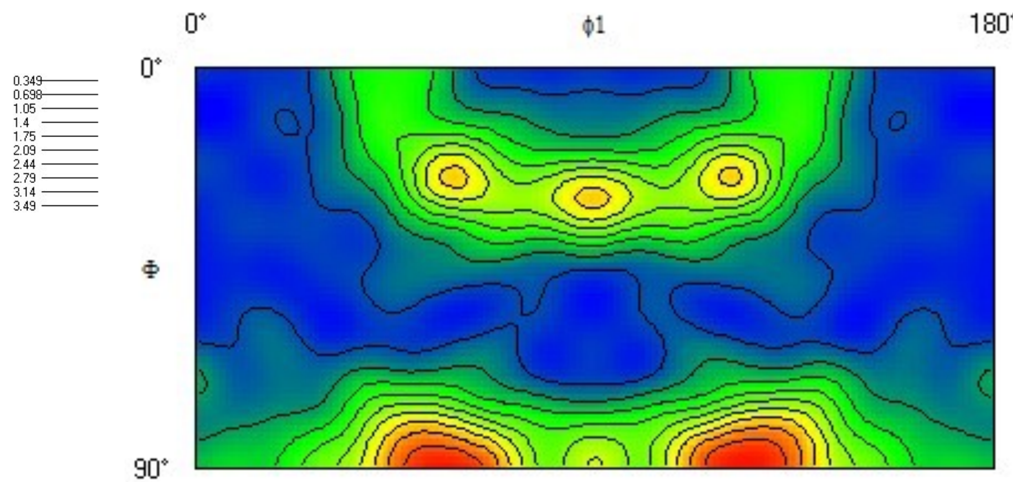


BATCH 2
as received

Goss {011}<100>
Brass {110}<112>

BATCH 2
1h at 930°C

Goss {011}<100>
Brass {110}<112>
Cube {001}<100>
Copper {110}<111>



Specimen Preparation

20Cr25Ni Nb-stabilised s.s.

- cold work
- 40min at 930°C

Annealed specimen

- irradiation

Irradiated specimen

Annealed specimen:

- austenitic matrix
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Irradiated specimen:

- austenitic matrix
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  - texture -> EBSD
- second phases
  - composition -> SEM & TEM
  - texture -> EBSD

Contents

Conclusions

- Proton irradiations of 20Cr25Ni Nb-stabilised s.s. can be performed in controlled conditions of damage and temperatures at DCF
- The heat treatment (40min at 930°C) produces a partially recrystallised austenitic structure with intra and inter-granular Nb-rich second phases
- The annealed austenitic matrix has a {101}<111> texture

Contents

Future Work

- Micro-structural characterisation of heat treated samples
- New irradiation experiment in January 2017
- Preparation of irradiated specimens for AEM using
  - electropolishing
  - (FIB) - > Site-specific specimen preparation
    - grain boundaries -> RIS
    - second phases -> precipitates stability under irradiation

Thank you for your attention

Questions?

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EPSRC Engineering and Physical Sciences Research Council

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[2] D.I.R. Norris,C. Baker, J.M. Titchmarsh, Radiation-induced sensitisation of stainless steel, Berkeley Nuclear Laboratories, 1986.
[3] P. T. Wady, A. Draude, S. M. Shubeita, A. D. Smith, N. Mason, S. M. Pimblott, E. Jimenez-Melero, Nucl. Instr. Meth. Phys. Res. A 806 (2016) 109.

Investigations into the formation of Uranium Hydride

James Ed Darnbrough

DISTINCTIVE Theme 1 meeting

Penrith

15/11/16

Talk Outline

- Interest
- Bulk investigation
- In-situ Synchrotron
- STEM and EELS study
- Conclusions

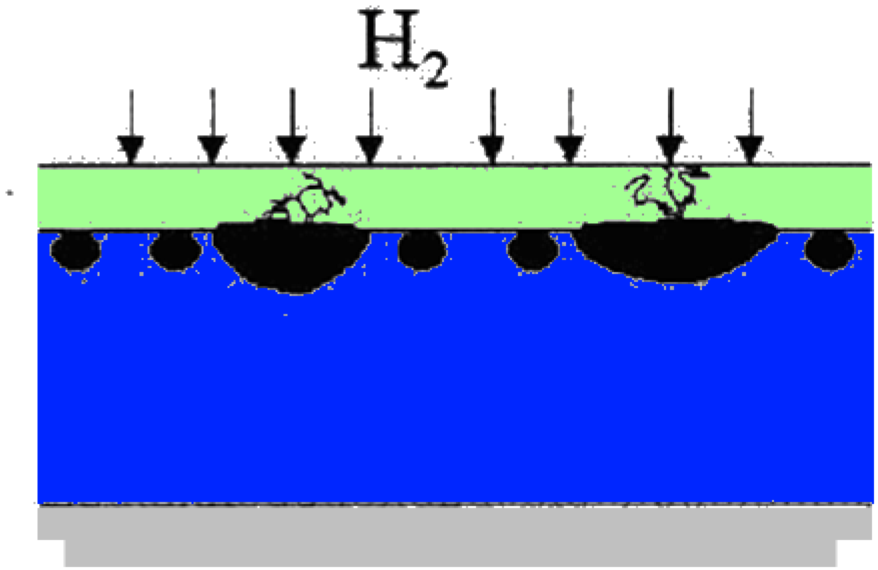
Interest in Uranium metal corrosion

- Legacy uranium fuels
  - Magnox fuel (UK),
- High density metal fuels (Advanced Test Reactors)
  - Uranium Aluminium, Uranium Zirconium, Uranium Silicon, Uranium Molybdenum
- Analogous to Pu

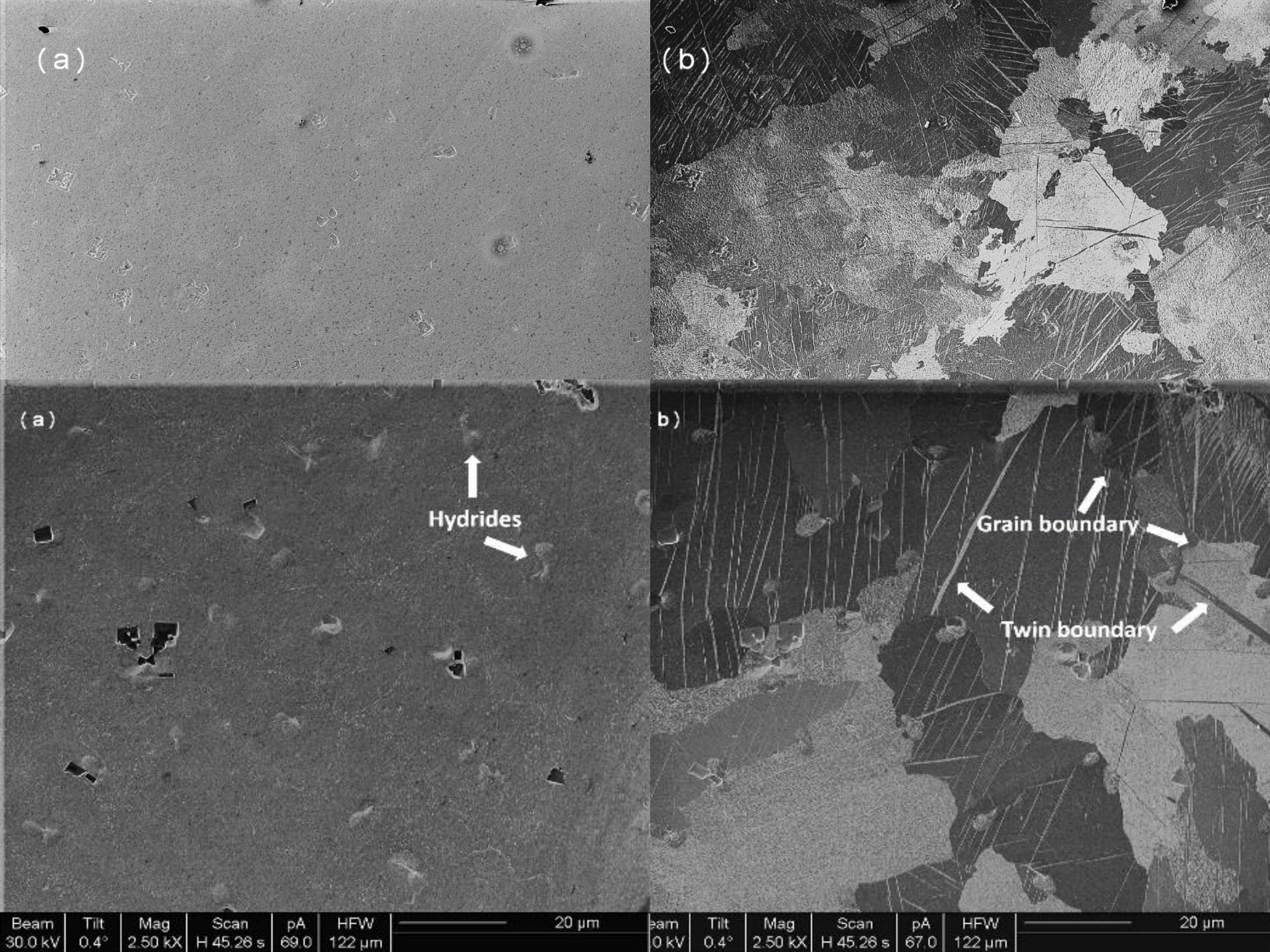
Uranium Hydride

Proposed Hydride Formation

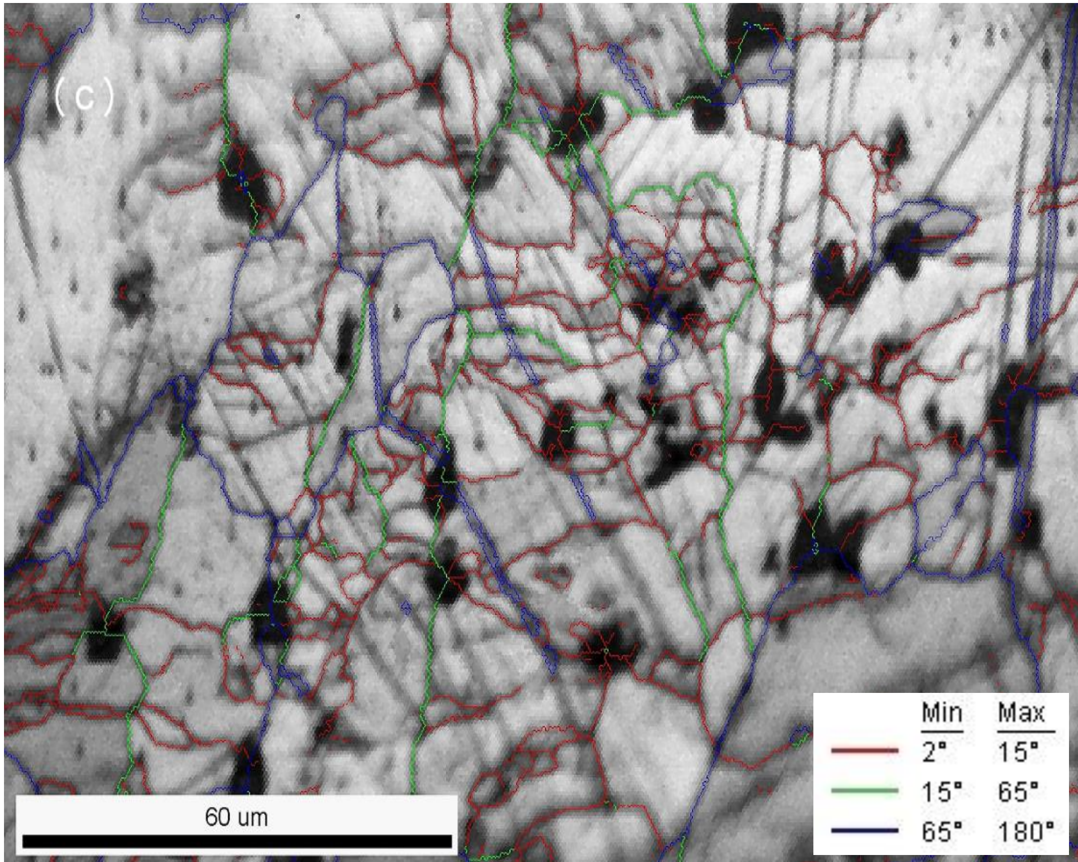
$2U + 3H_2 \rightarrow 2UH_3$



Y. Ben-Eliyahu, M. Brill, M. H. Mintz, *J. of Chemical Physics*, 111 (1999) 6053-6060

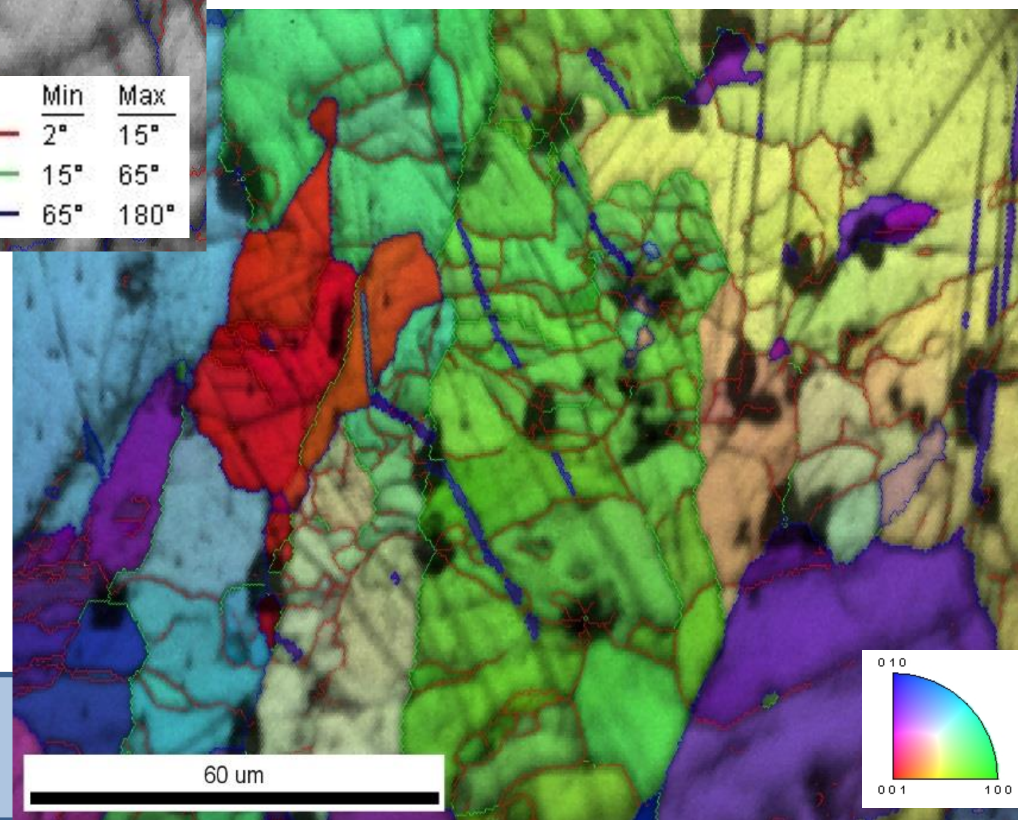


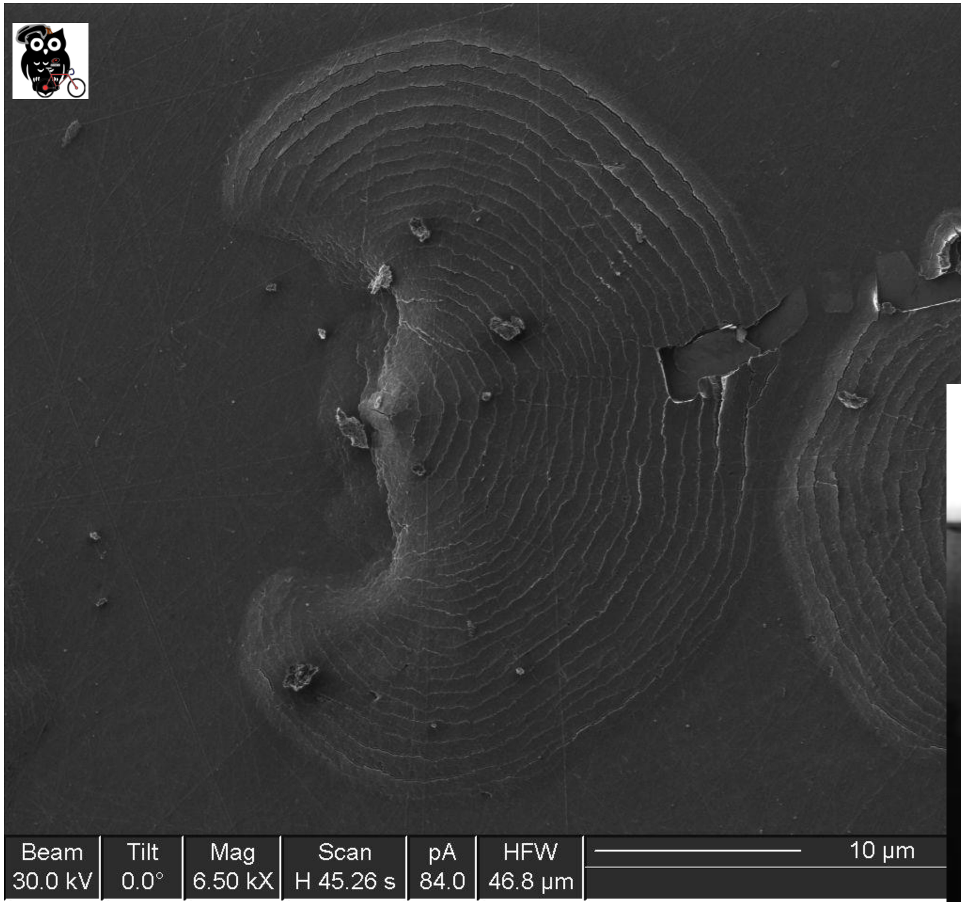
(a)
(b)
(a)
Hydrides
b)
Grain boundary
Twin boundary
Beam 30.0 kV
Tilt 0.4°
Mag 2.50 kX
Scan H 45.26 s
pA 69.0
HFW 122 µm
20 µm
Tilt 0.4°
Mag 2.50 kX
Scan H 45.26 s
pA 67.0
HFW 122 µm
20 µm

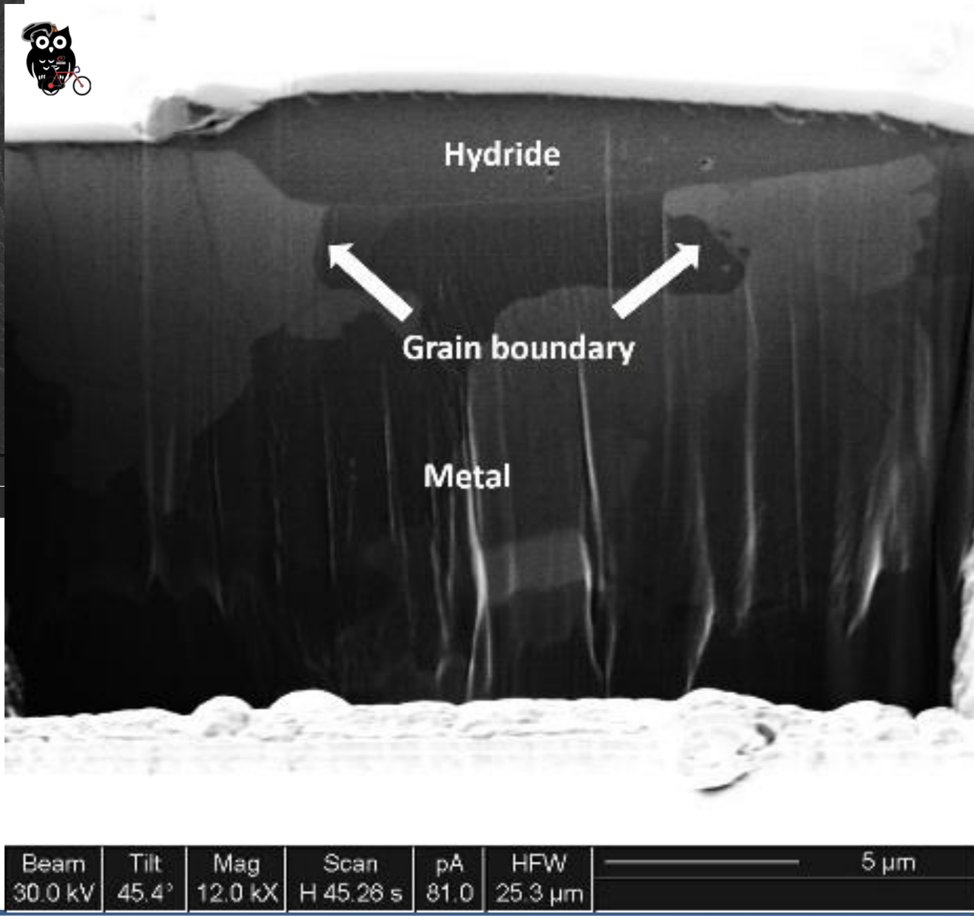


Blisters seen at triple points

Blisters seen at high angle boundaries







U 19.1 g/cm^3

↘

UH_3 10.95 g/cm^3

Significance of Blister Location

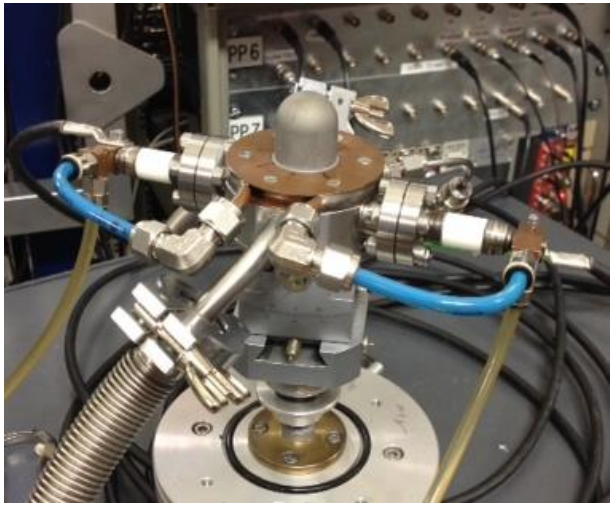
- At Triple points and high angle grain boundaries
- High diffusion paths?
- Free surfaces with lower density than crystalline material?
- Preferred growth direction?
- Where does it start in uranium metal?

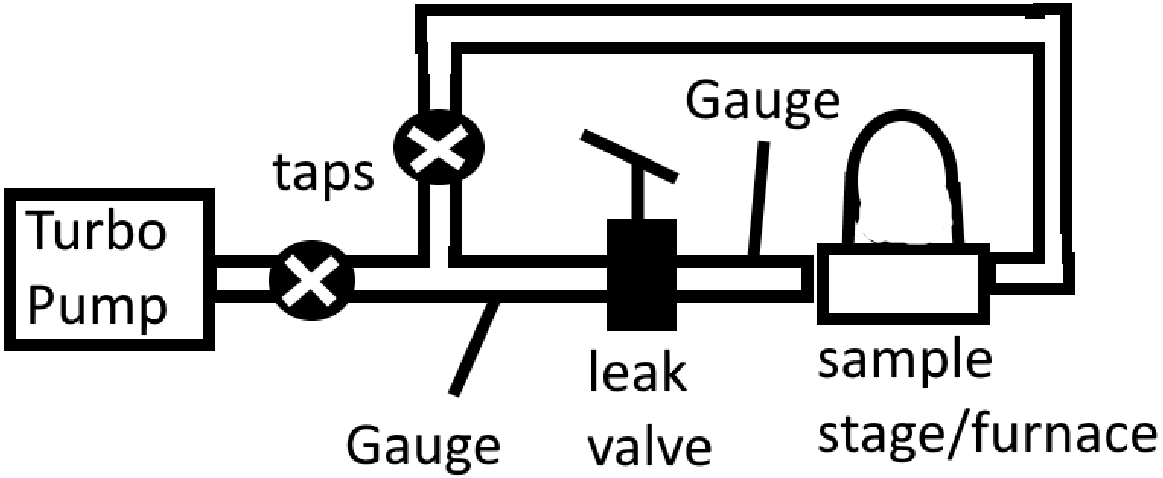
in-situ experiment

- Thin film samples produced (by DC magnetron sputtering) to mimic interface without large masses reducing the activity to exempt limits
- Samples are produce a uniform homogeneous oxide thickness on a known metal thickness
- Utilised high angle diffraction to look at crystallinity and low angle reflectivity to look at layer interfaces

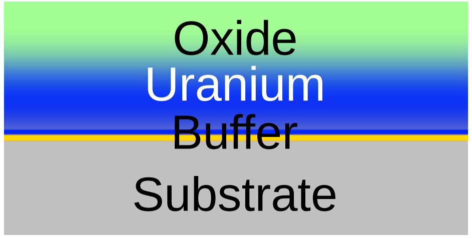
Experimental set up

- Used XMaS beamline at ESRF
- Furnace mounted on goniometer allowed remote temperature control (RT-200°C) and gas dosing (1000mbar of 4%H_2/Ar)



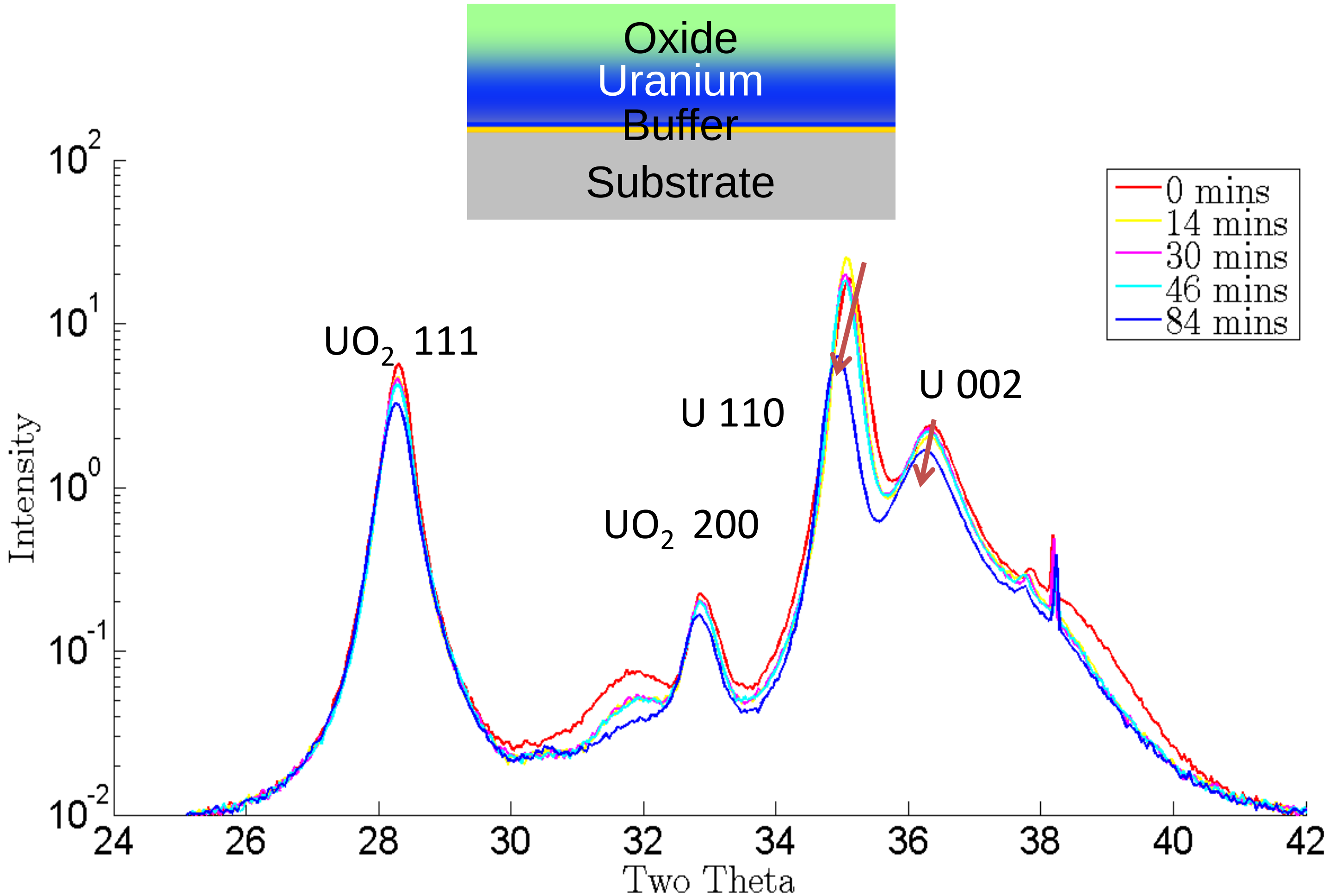


Introduction of H_2

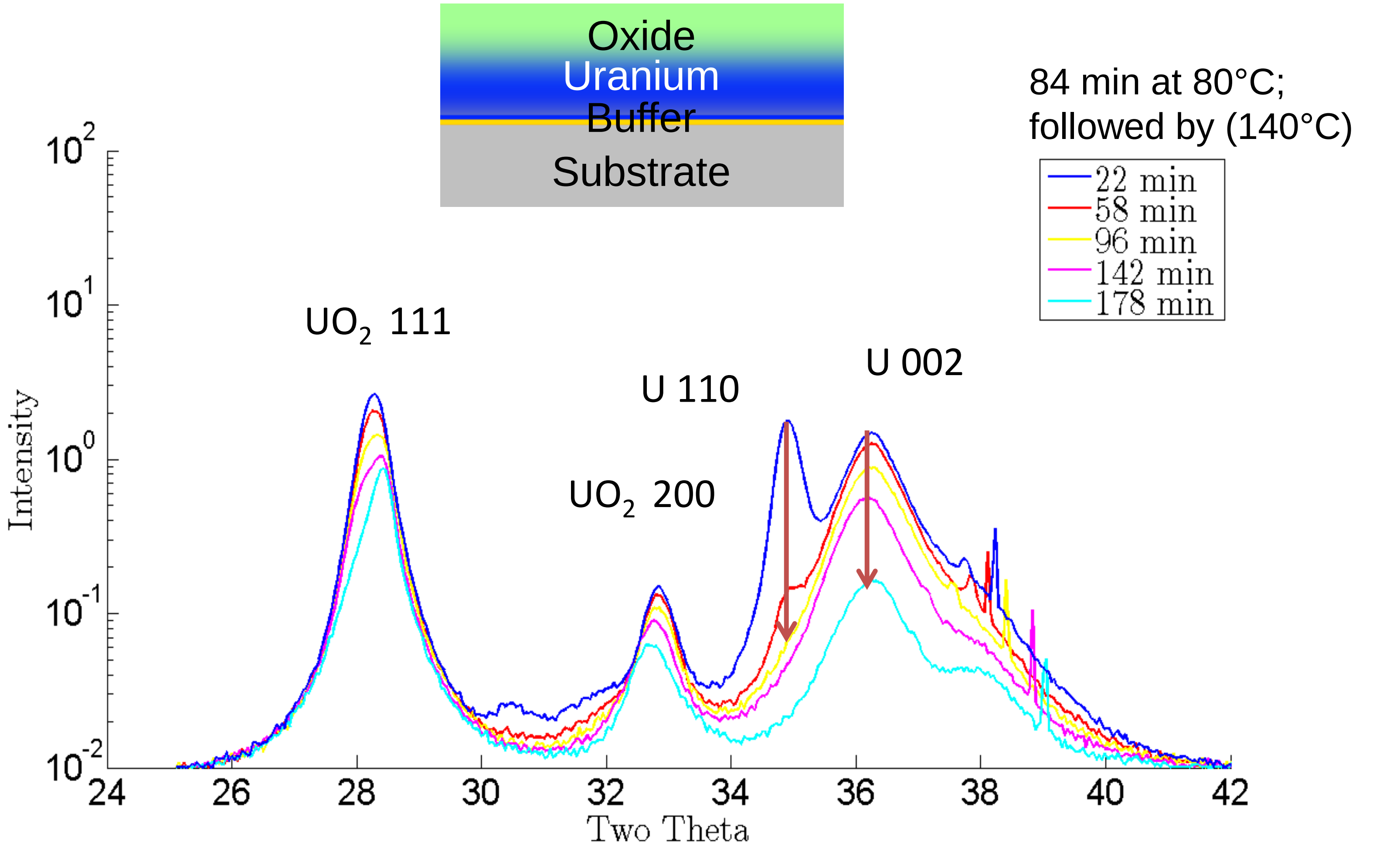


- U main orientation (110)
- U (002) intensity at approximately 10% of (110)
- UO_2 more intense predominantly (111) with some (200); higher UO_{2+x} oxides also observable
- Hydrogen Exposure
  - 84min 50 mbar gas mix (2 mbar H_2) at 80°C
  - Further 187min at 400 mbar gas mix (16 mbar H_2) at 140°C
  - And 156min 400 mbar gas mix (16 mbar H_2) at 200°C

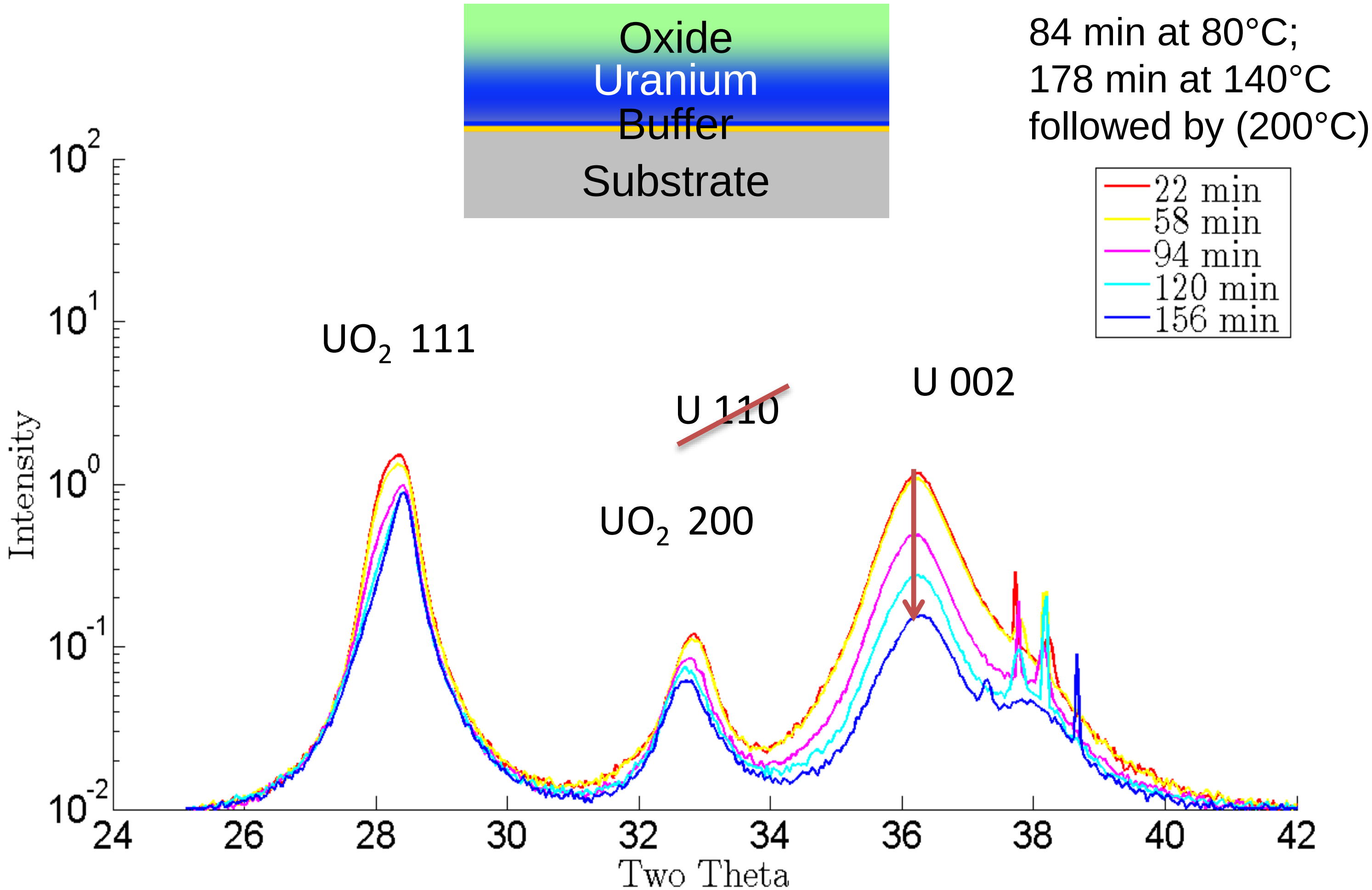
80 °C exposure



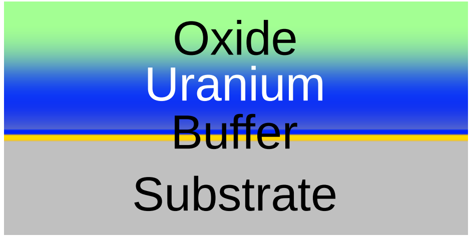
140 °C exposure

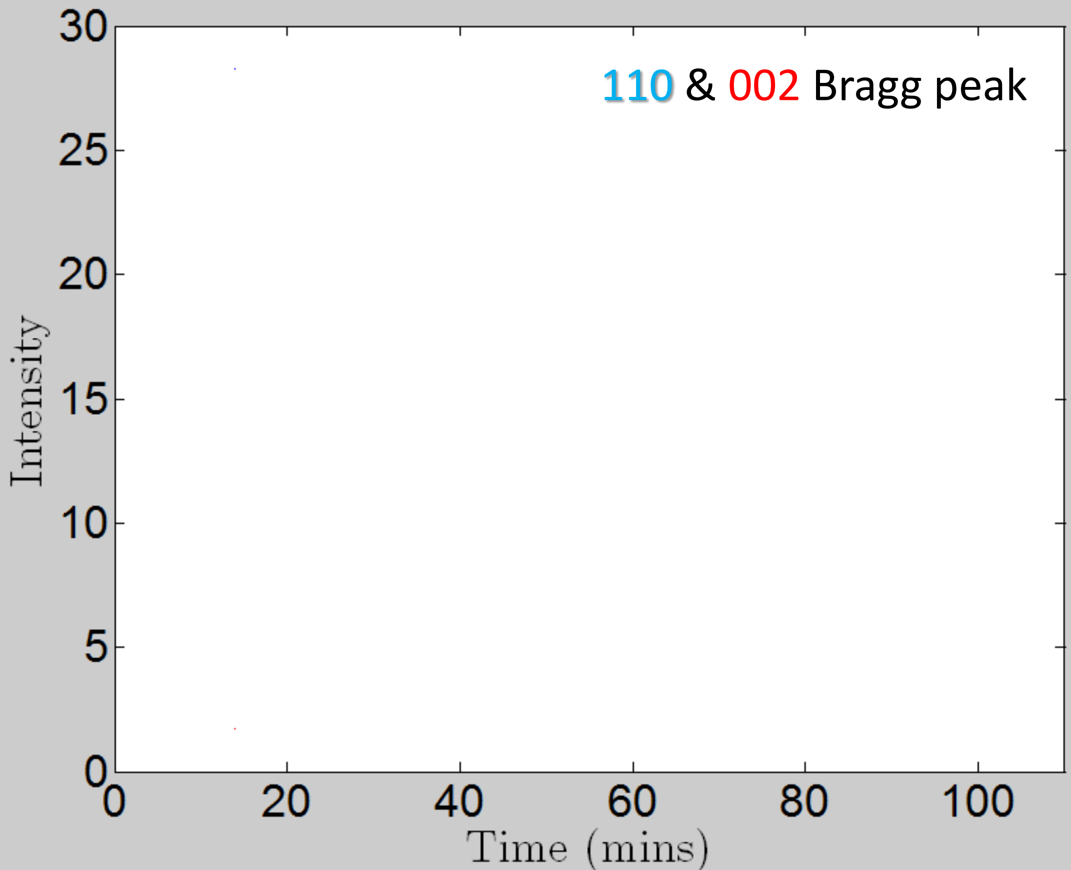


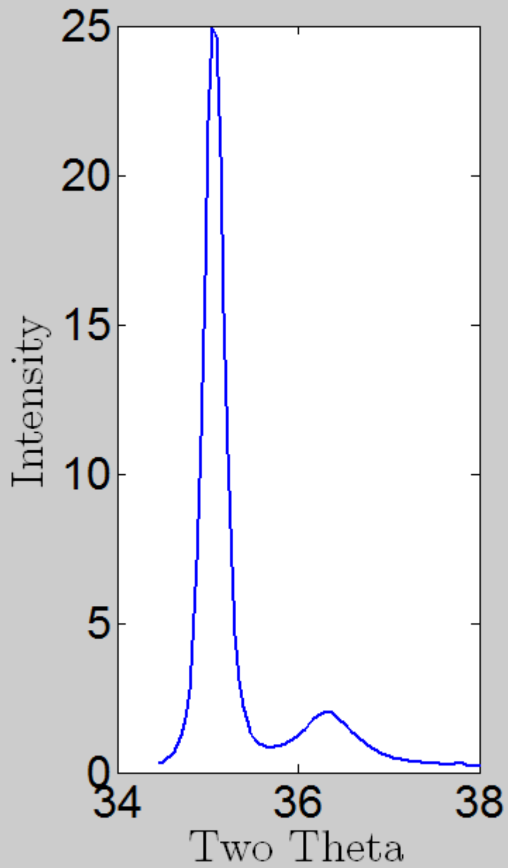
200 °C exposure



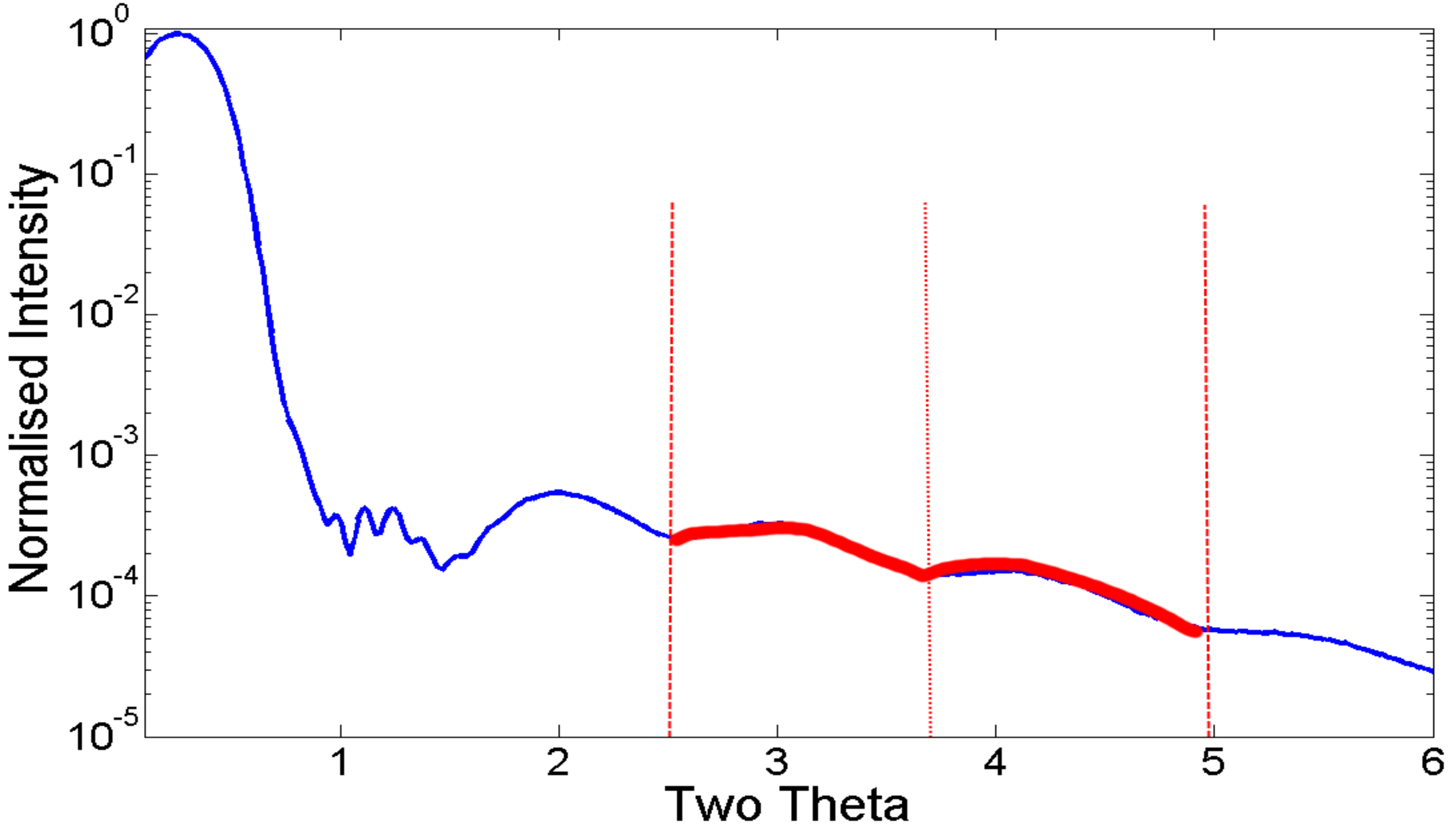
Rate of Crystalline Removal



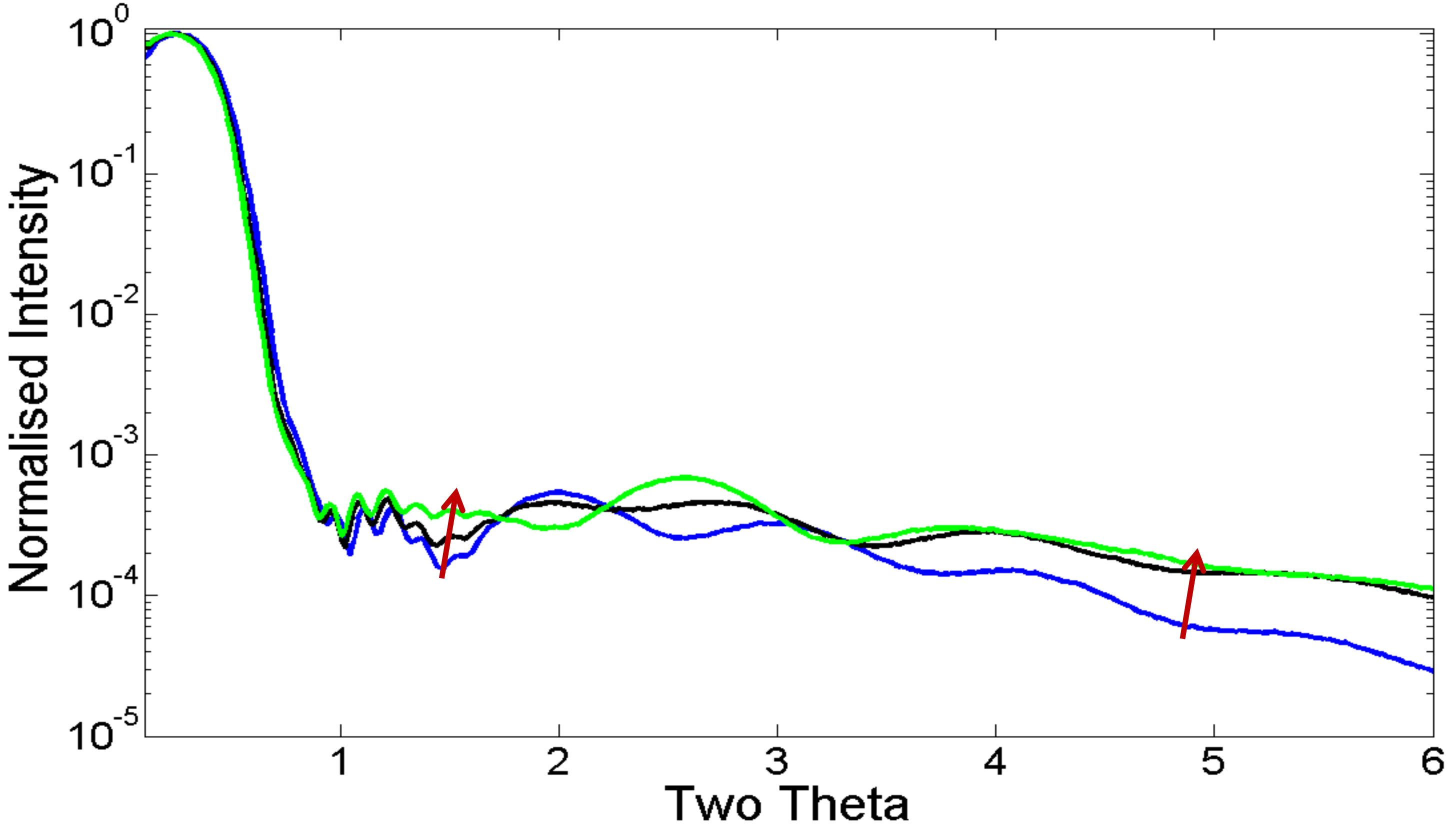




Reflectivity



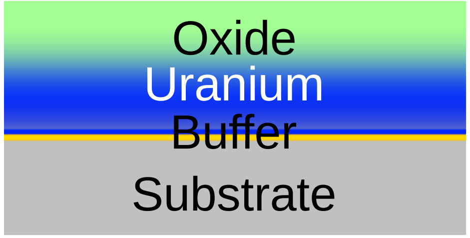
Reflectivity

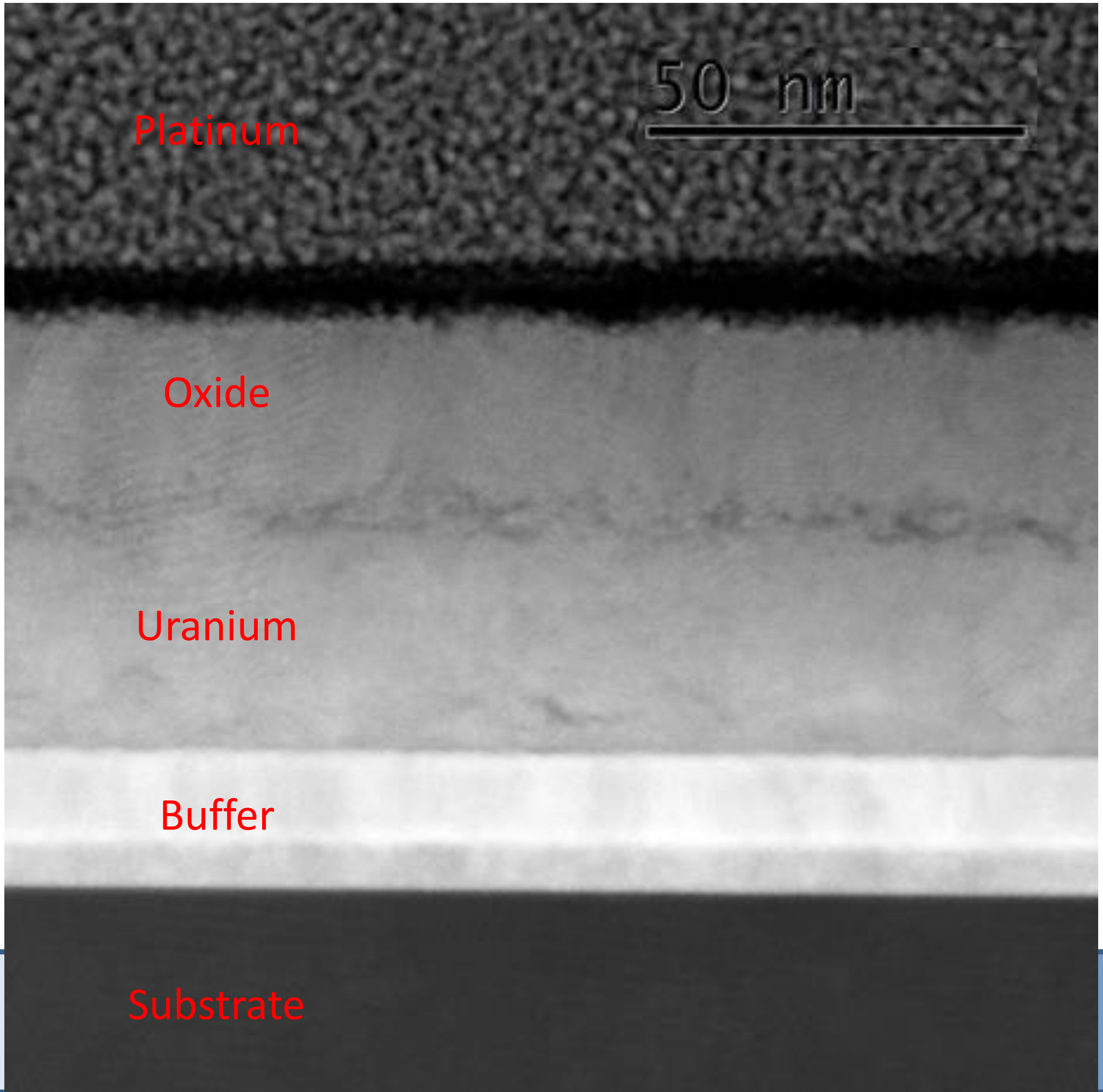


X-ray Findings

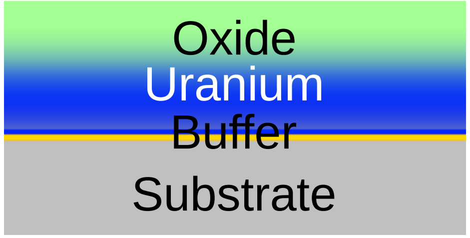
- No identification of crystalline β-UH_3 (seen in bulk U experiments under similar conditions)
- Consumption of U metal (2 mbar P_{H2}, 80-140°C) monitored in-situ by synchrotron XRD
- Preference of consumption of (110) over (002) demonstrated
- Oxide consumed and strained throughout with higher oxides (UO_{2+x}) also reduced
- Reflectivity shows reduction in U metal density material
- Increase in amount of UO_2 (10.97g/cm^3) or UH_3 (10.95 g/cm^3) density material
- Roughening of the U/UO_2 interface

STEM Bright Field

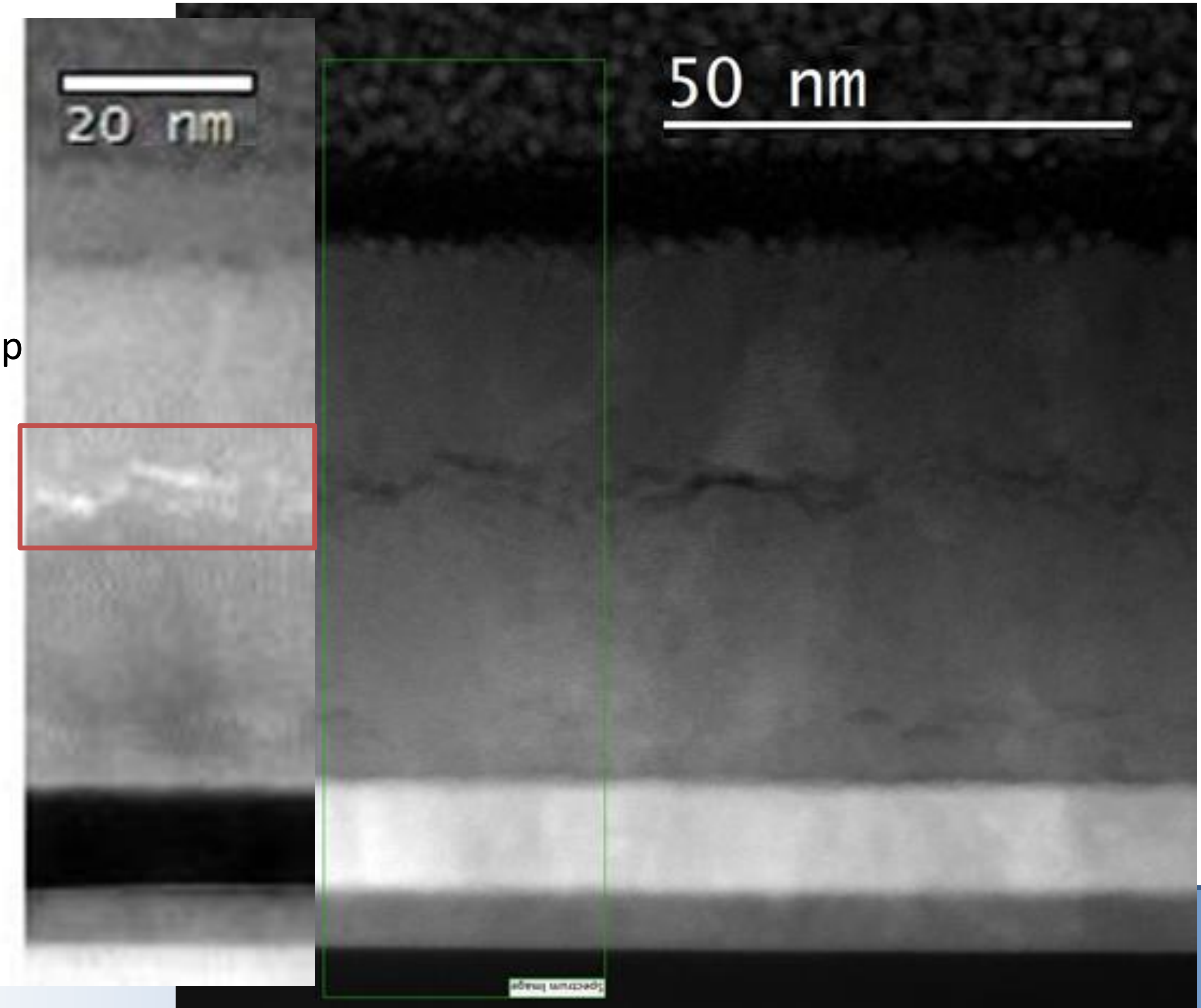




STEM
High Annular Dark Field



EELS
Selected
Peak map



Hydriding of Uranium

- Small partial pressure of H_2 and moderate temperatures over hours forms hydride
- Different Uranium crystallographic directions are preferentially consumed
- Amorphous hydride produced at the interface between U and UO_2
- No crystalline hydride observed

Acknowledgements

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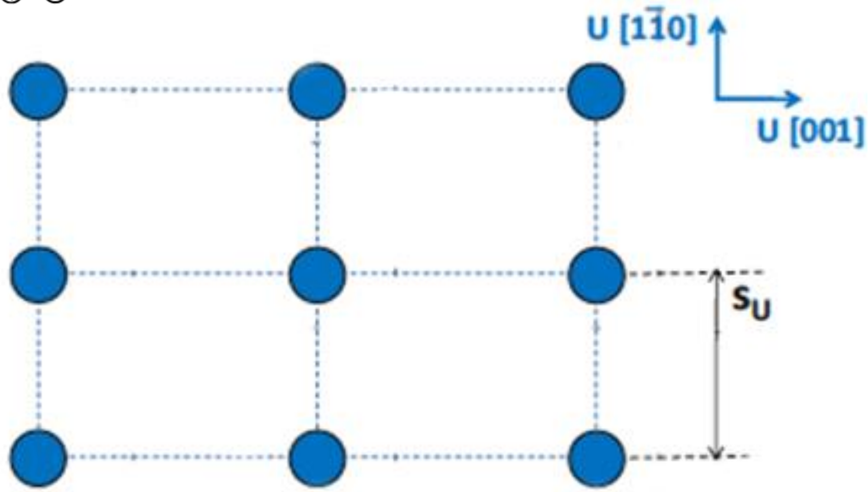
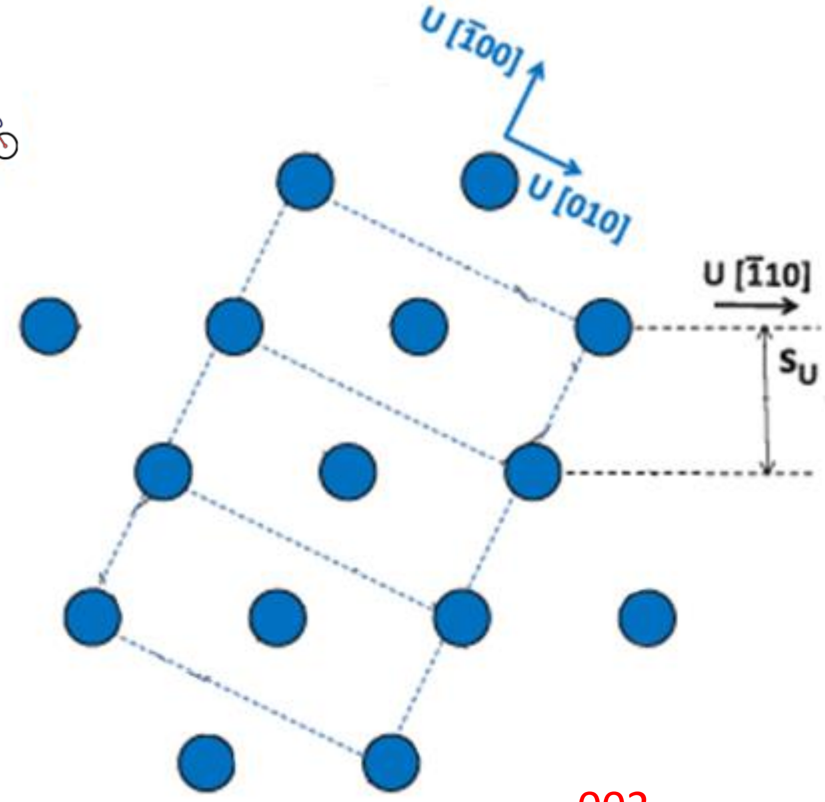
  - Ross Springell
  - Tom Scott

Crystallographic Dependence



110



002