Studsvik

Microstructural and Chemical Characterization of Irradiation Effects in Claddings with Different Alloy Compositions

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INTRODUCTION

- Four TVEL fuel segments were transported to Studsvik Hot Cell Labs (HCL) from the Halden HBWR during 2018

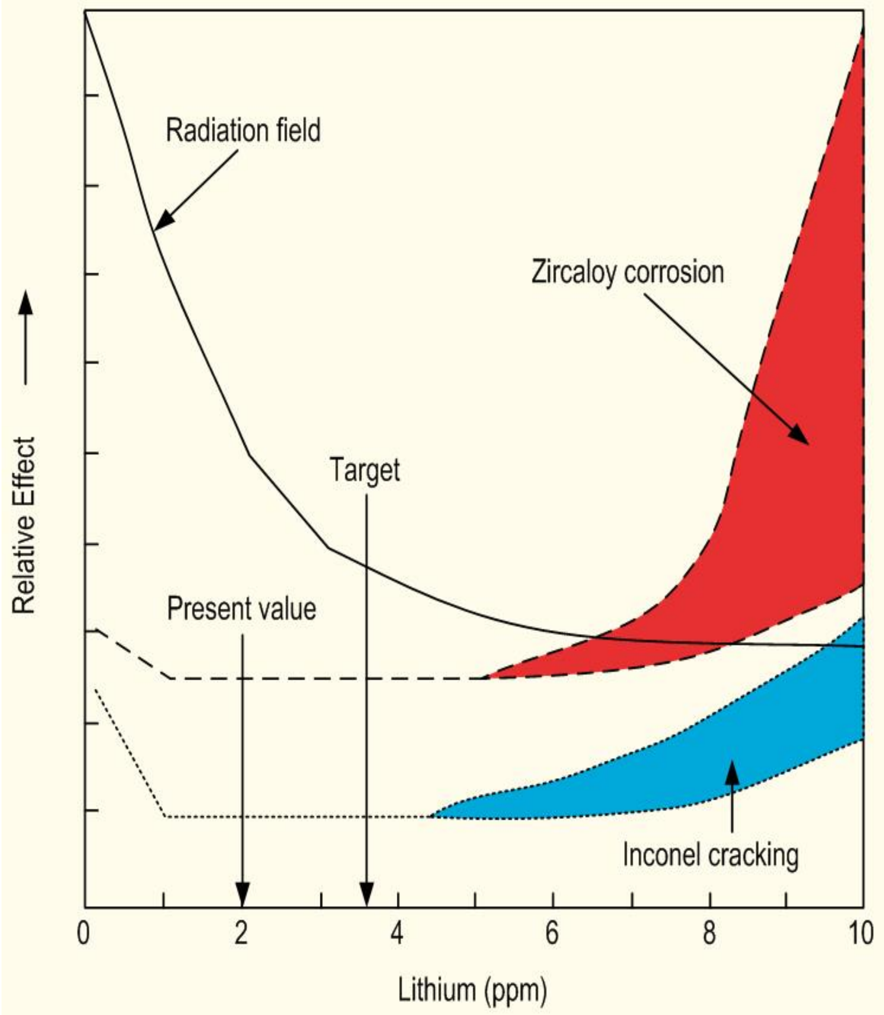
- The segments were irradiated as a part of the IFA.728

- An extensive scope of “advanced PIE” has been performed, including high resolution scanning electron microscopy (SEM), transmission electron microscopy (TEM), and laser ablation inductively coupled plasma mass spectroscopy (LA-ICP-MS)

- Main purpose of the examinations: to support the ongoing licensing of TVEL PWR fuel in US, Europe and Asia, with a special focus on the in-pile response of TVEL claddings in elevated Li PWR water chemistry

- The PIE program scope and preliminary results will be presented

BACKGROUND



"PWR Primary Water Chemistry Guidelines," EPRI 1999

- Lithium and Boron in Ringhals 3
  - $^{7}LiOH$ added to PWR coolant @ BOC ⇒ neutralise primary water to counteract H_3BO_3 (boric acid) for reactivity control
  - R3 runs elevated Li, >3-5 ppm to reach pH_{300} 7,4
- Low Li leads to low pH ⇒ increased corrosion of core internals ⇒ elevated CRUD deposition, reduced performance, higher dose rates
- Accelerated/corrosion of cladding associated with higher integrated Li exposure ⇒ increased probability of fuel failure
- Four TVEL segments irradiated in *very high* Li chemistry (≤10ppm) for over 900 days ⇒ focus on understanding oxide formation and lithium uptake in materials

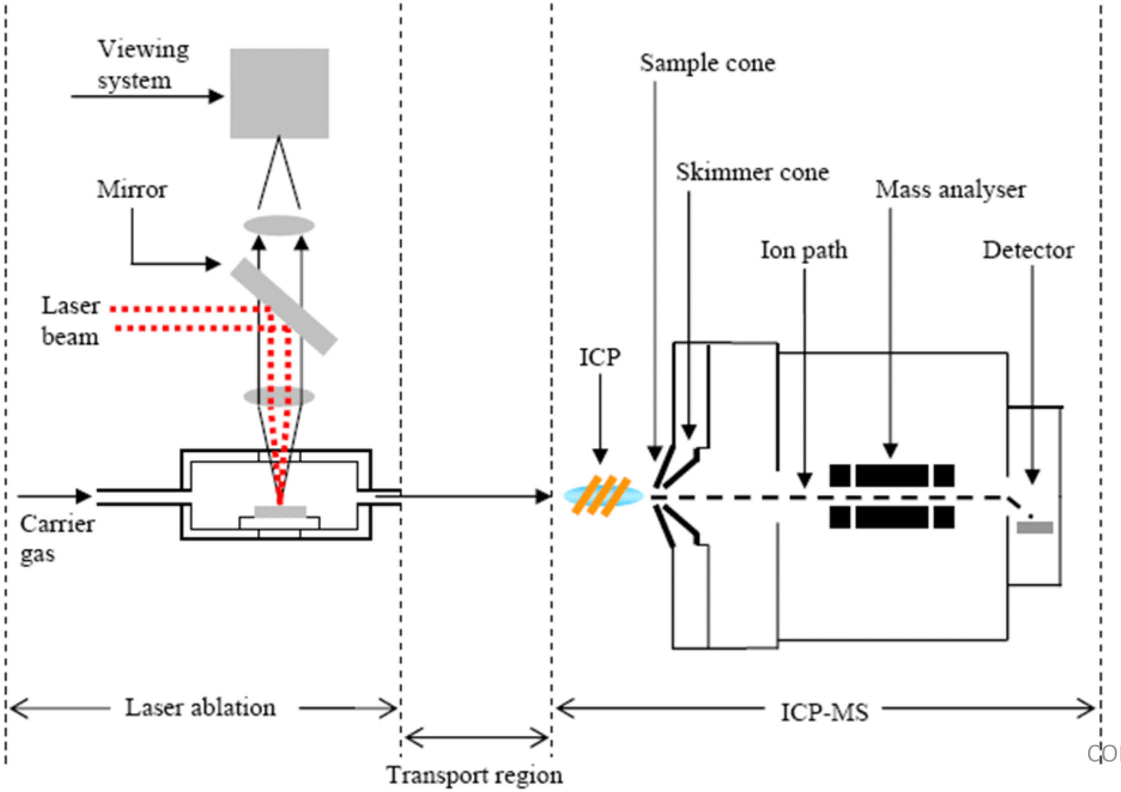
IRRADIATION AND MATERIAL DETAILS

System	Alloy	Nb	Sn	Fe	O	Oxide Thickness * (μm)
Zr-Nb-Sn-Fe	E635M	0.79	0.81	0.335	0.075	48.8
Zr-Nb	E110opt	1.05	-	0.055	0.085	18.3
	E110M	1.02	-	0.095	0.120	18.2
	E125	2.45	-	0.035	0.069	13.6

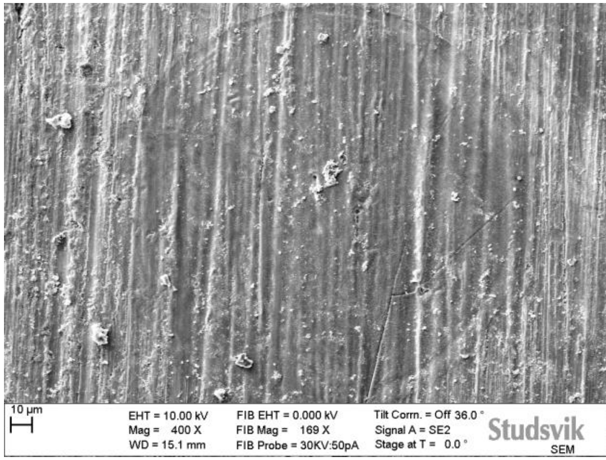
- The segments were irradiated at the Halden HBWR
  - E110opt – 47,3 MWd/kgUO_2
  - E110M – 47,8 MWd/kgUO_2
  - E125 – 45,5 MWd/kgUO_2
  - E635M – 45,7 MWd/kgUO_2
- Segments irradiated at 351°C and ≤10ppm Li RWC for 907 days
- NDE, LOM, and hydrogen measurements performed at the IFE Hot Cell facility in Kjeller, NO

LASER ABLATION EXAMINATIONS

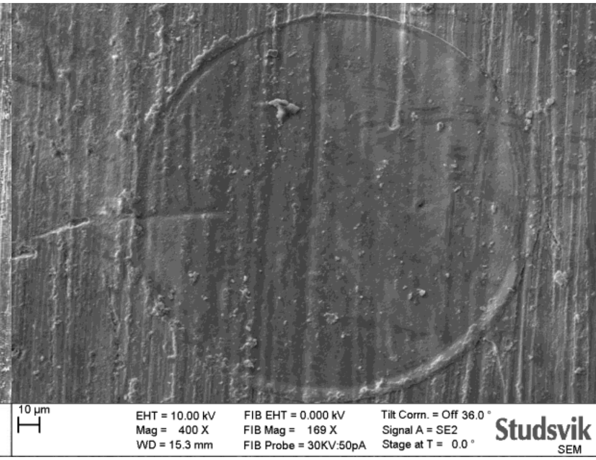
- Laser ablation on clad outer surface conducted, ablation productions fed through inductively coupled plasma mass spectrometer
  - Enables quantification of isotopes with standard
  - Focus on Li and B10/B11 distribution in oxide, qualitative analysis of Nb, alloying element composition
- Examinations and analysis currently ongoing



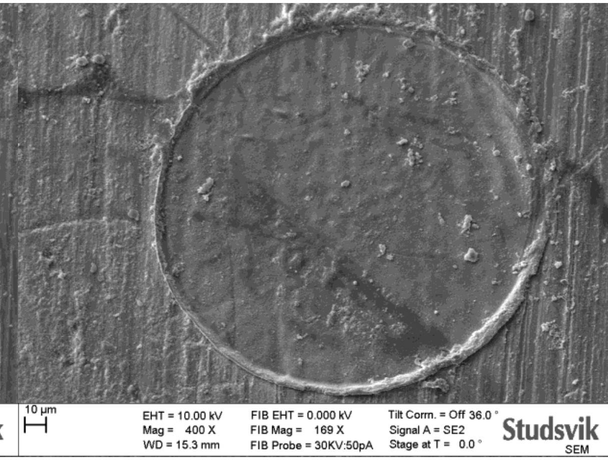
LASER ABLATION EXAMINATIONS



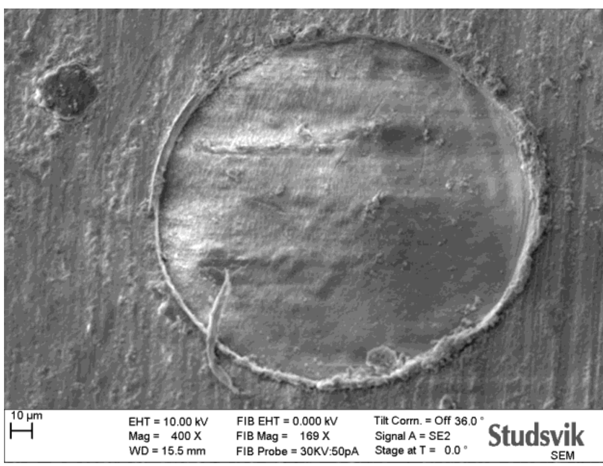
≈0.3µm



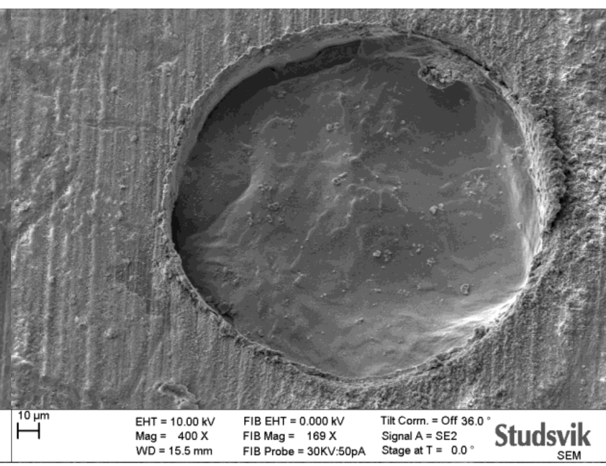
≈1µm



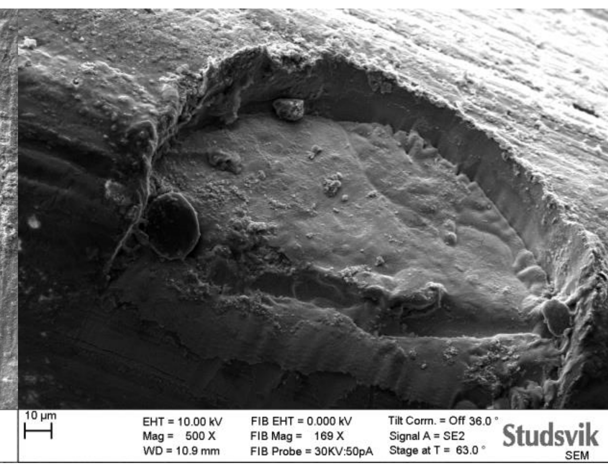
≈2.5µm



≈7µm

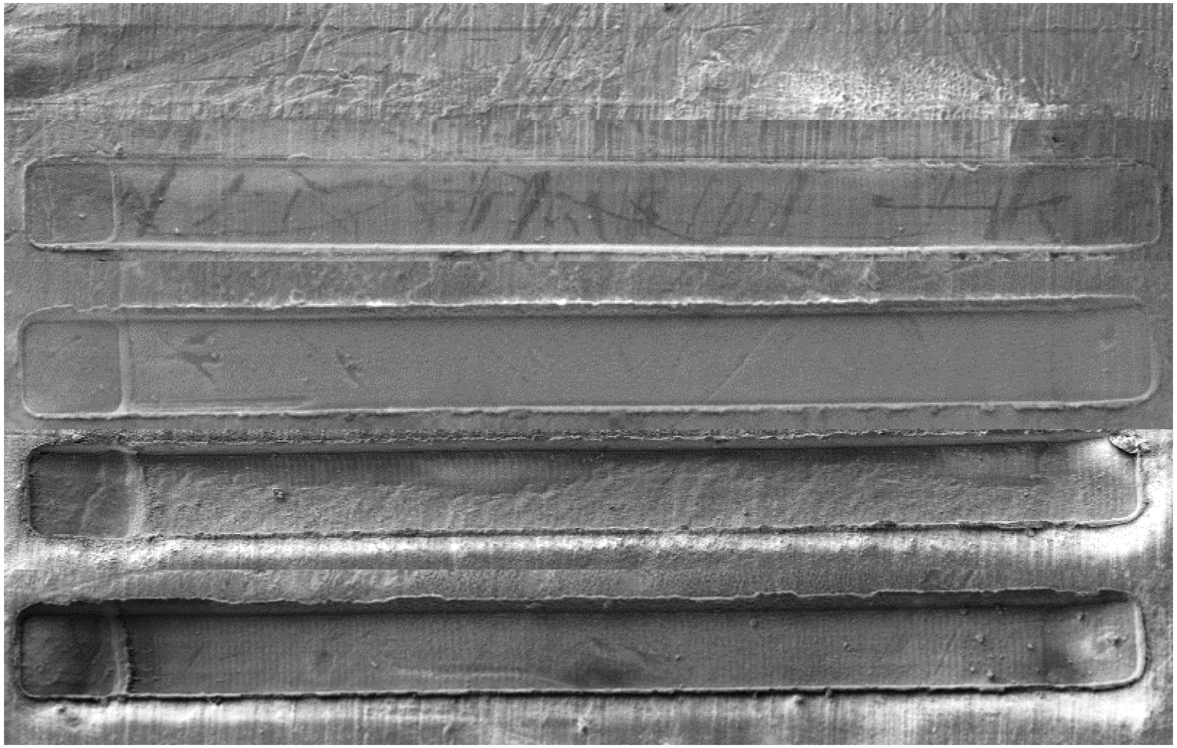


≈24µm



≈24µm

LASER ABLATION EXAMINATIONS



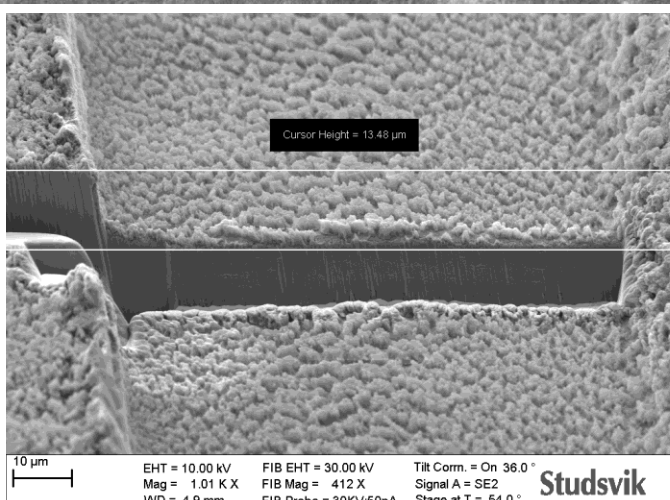
≈1µm

≈6µm

≈12µm

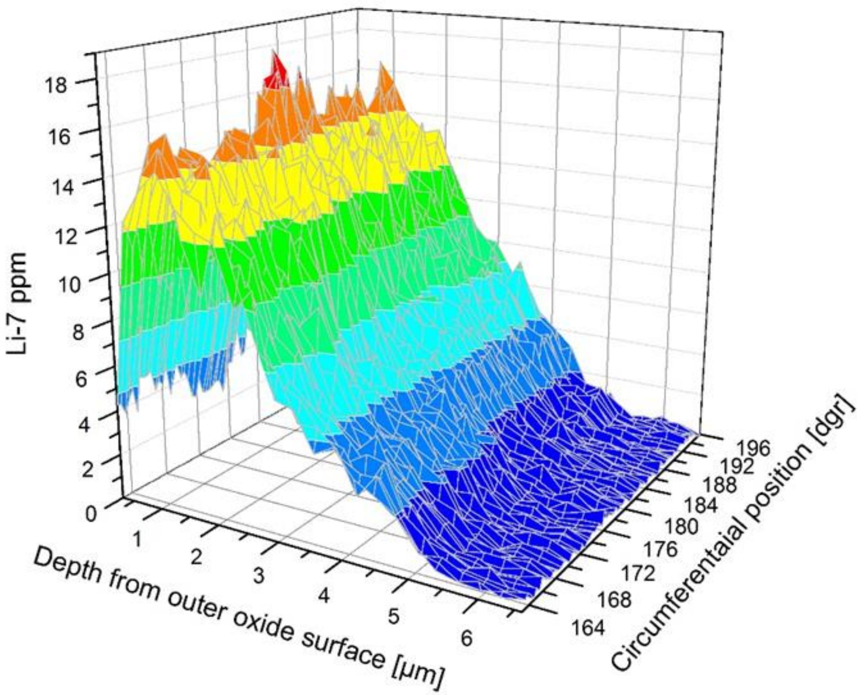
≈18µm

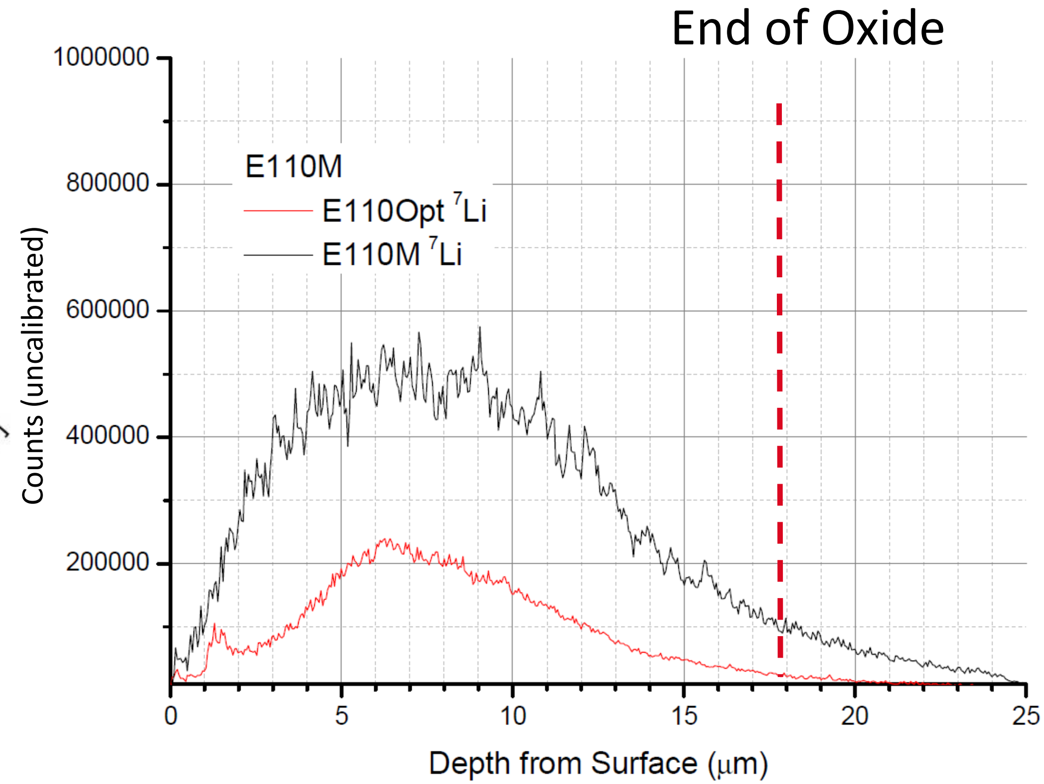
≈24µm



- Different scan types used to evaluate local isotopic concentration profile with high resolution (spot) as well as trends over larger spatial volumes (lines)

LASER ABLATION EXAMINATIONS





Typical results (left) from zry clads run in elevated lithium water chemistry in power reactors compared to preliminary results from E110M and E110-Opt run in very high Li in HBWR (right), indicating higher Li uptake in E110M

A. Puranen et al, LITHIUM AND BORON ANALYSIS BY LA-ICP-MS RESULTS FROM A BOWED PWR ROD WITH CONTACT, Proceedings of Top Fuel 2015

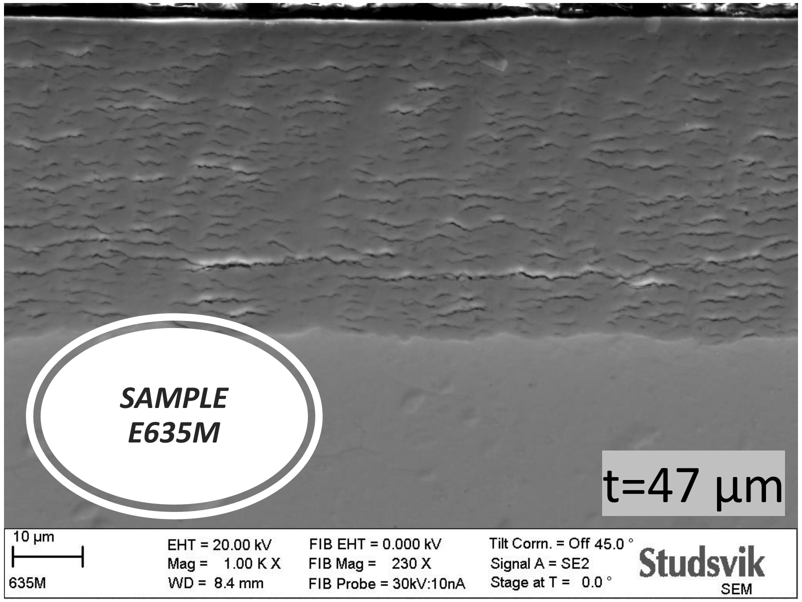
SCANNING ELECTRON MICROSCOPY EXAMINATIONS

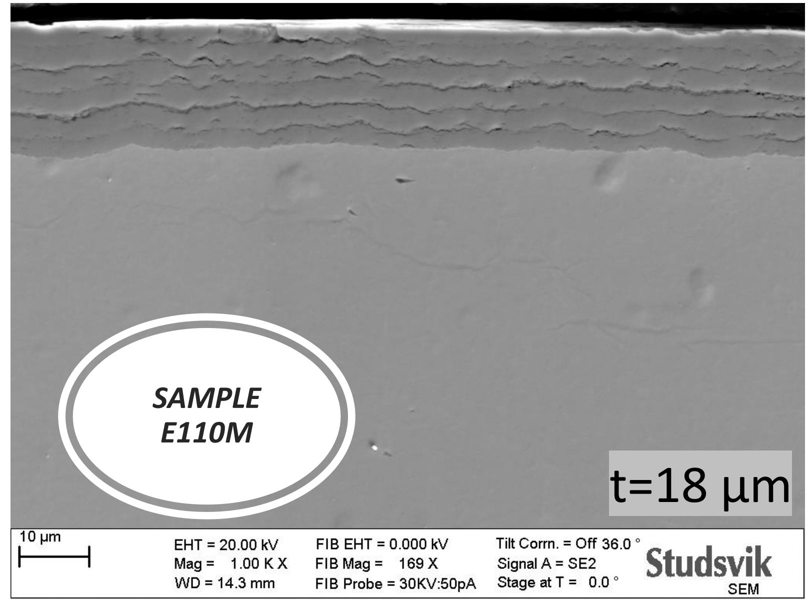
- A Zeiss Auriga cross beam instrument with EDS, WDS and EBSD was used for the SEM work and the FIB lift-outs for TEM analysis.
- Cross section samples were prepared using standard metallographic techniques.

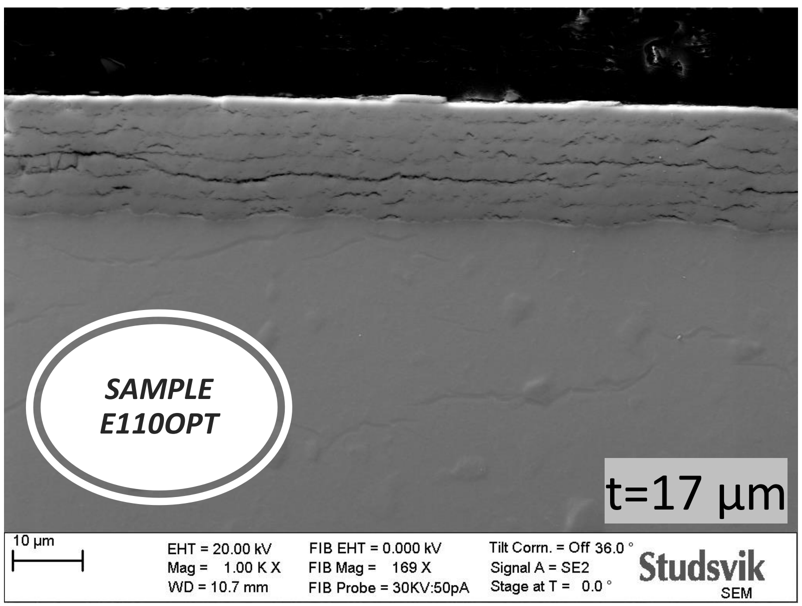
- SEM was used to characterize the oxide layer morphology and composition as well as the metal-ocide interface region.
  - Alloying element distribution?
  - Any impurities present?

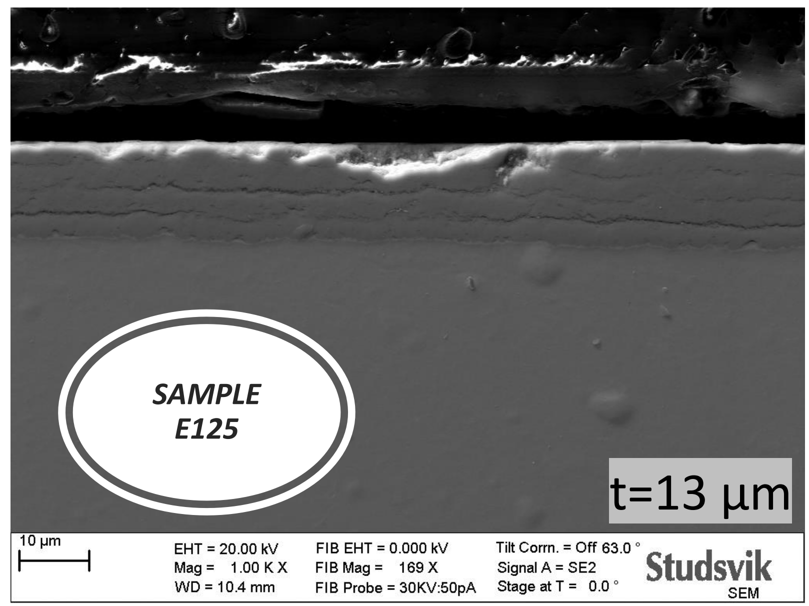
- The hydride morphology was also characterized.

OXIDE THICKNESS AND MORPHOLOGY (1)

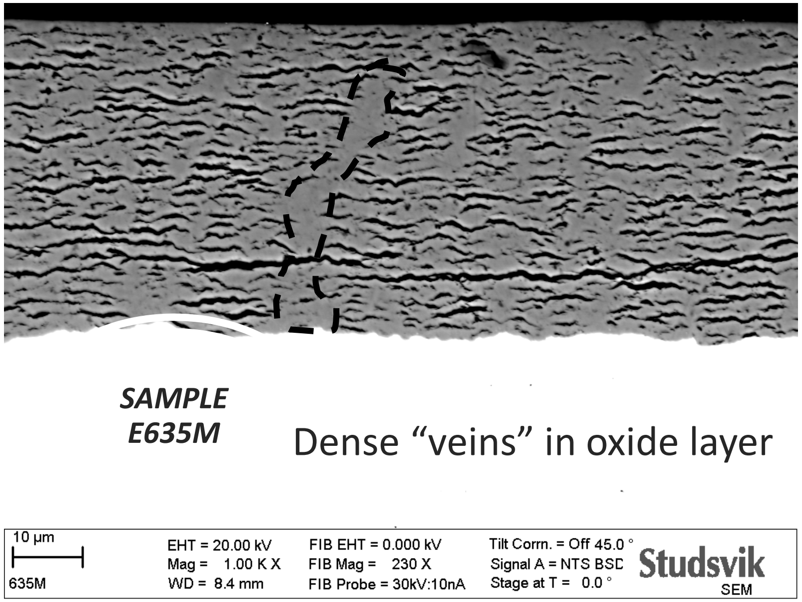






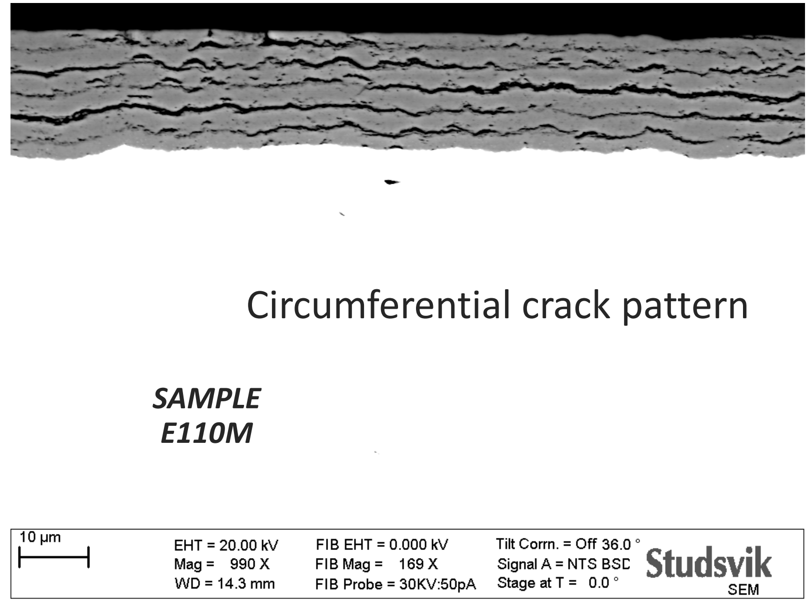


OXIDE THICKNESS AND MORPHOLOGY (2)



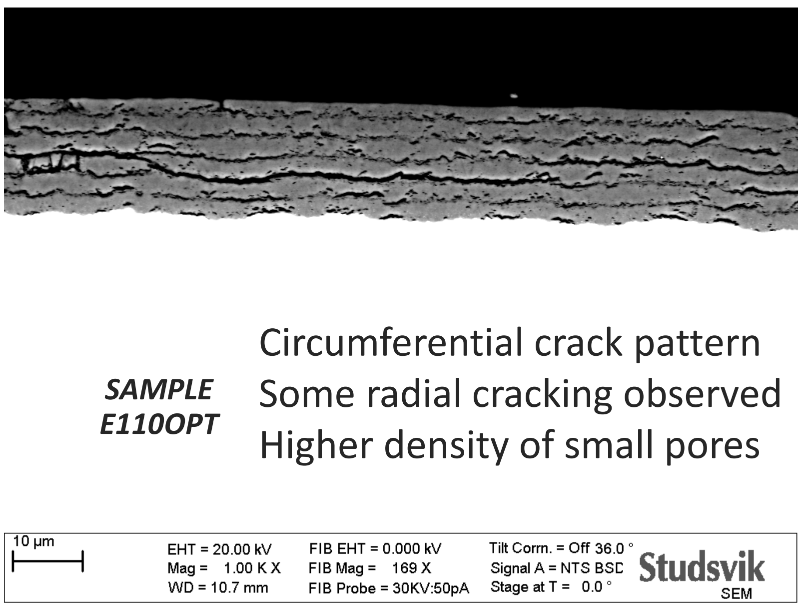
SAMPLE E635M

Dense “veins” in oxide layer



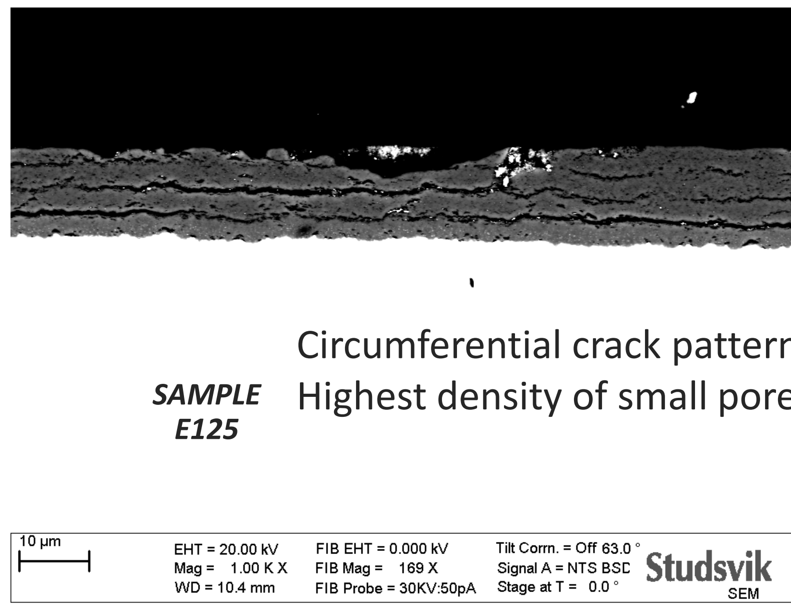
SAMPLE E110M

Circumferential crack pattern



SAMPLE E110OPT

Circumferential crack pattern
Some radial cracking observed
Higher density of small pores



SAMPLE E125

Circumferential crack pattern
Highest density of small pores

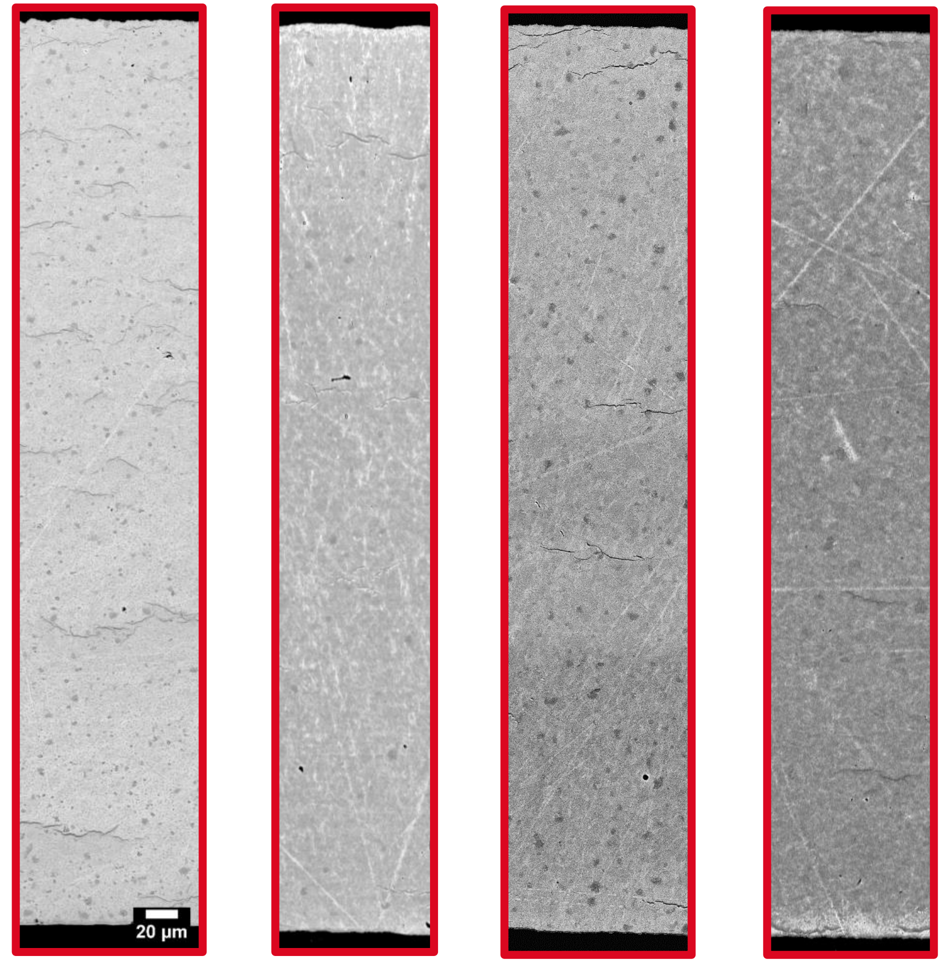
HYDRIDE MORPHOLOGY

- Hydrides evenly distributed throughout the cladding – no rim formation at these low hydrogen concentrations
- More hydrogen in the E635M alloy correlating well to the thicker oxide layer
- Very low hydrogen concentrations in the other alloys

Water facing side

Sample E635M

Sample E110opt

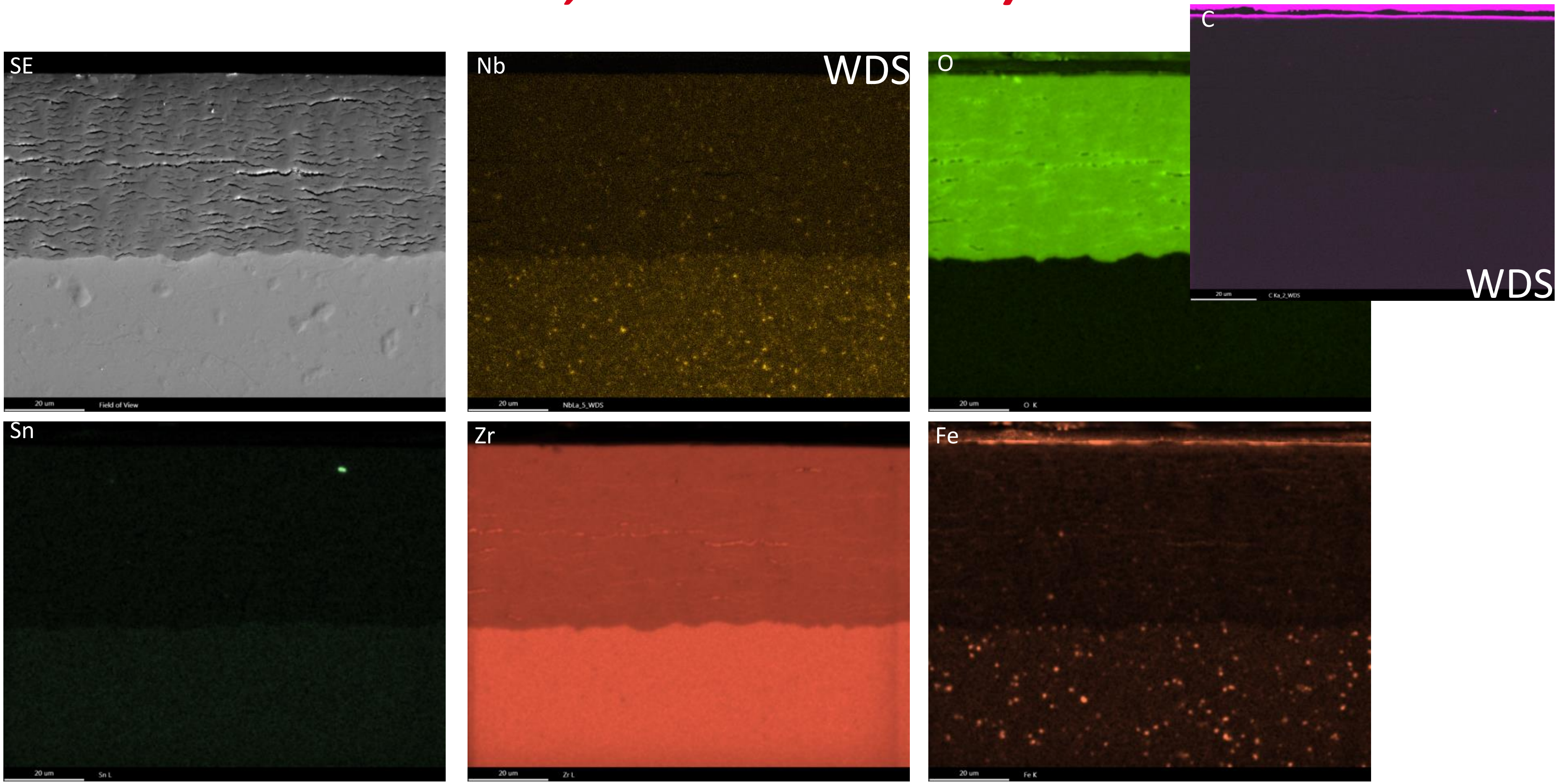


Sample E110M

Sample E125

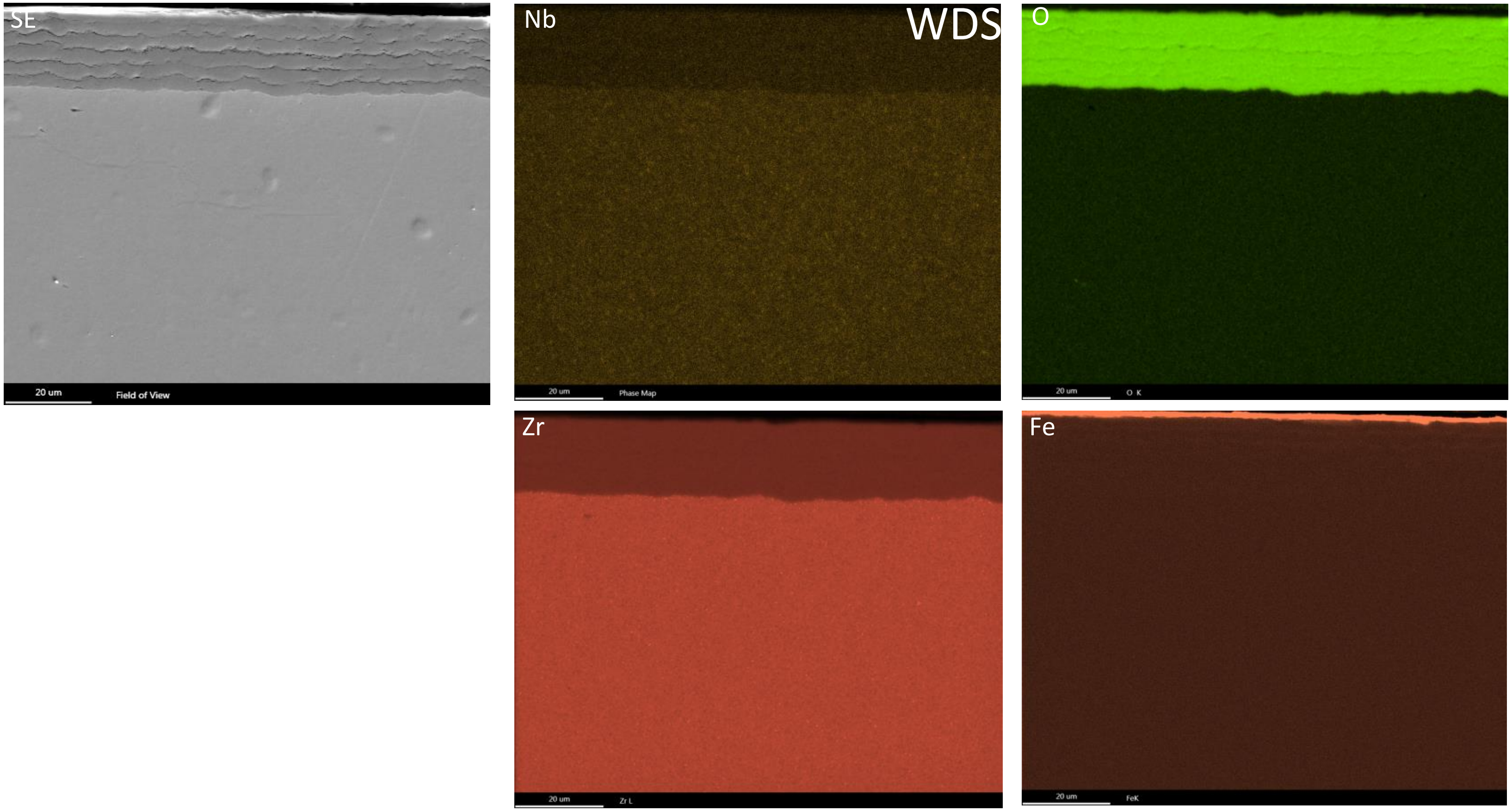
Fuel facing side

SAMPLE E635M – NB, SN & HIGH FE – E/WDS MAPPING



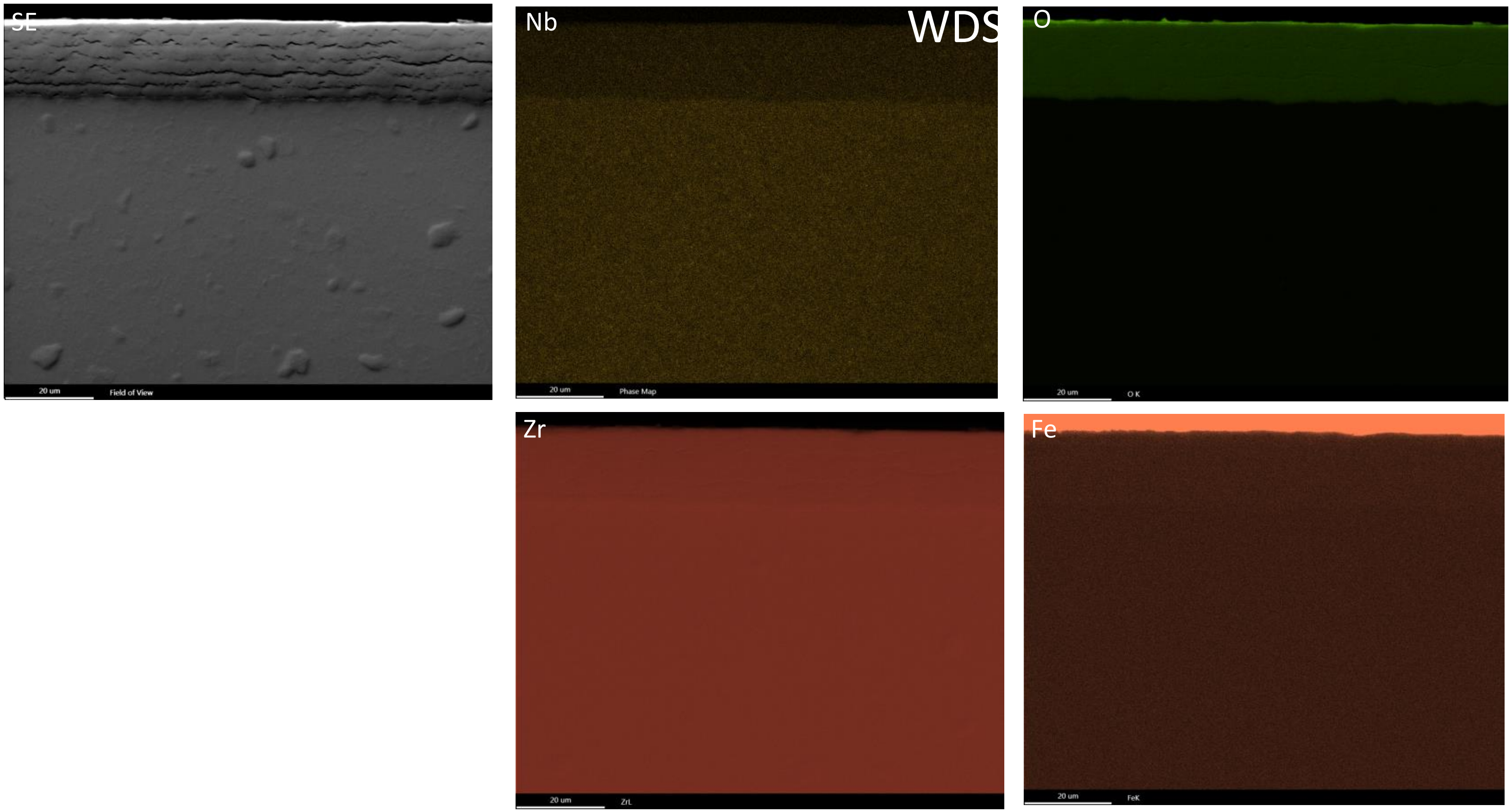
- Relatively large particles in this material easily detected using EDS and WDS
- No major loss of Fe to the matrix phase can be observed from these data (as is usually observed on similar materials). Most particles seem to have Zr-Nb-Fe compositions
- Interestingly, particles seem to dissolve in the oxide. The further out in the oxide the fewer and faint particles. In the outer oxide no particles can be observed.

SAMPLE E110M – NB & "HIGH" FE – E/WDS MAPPING



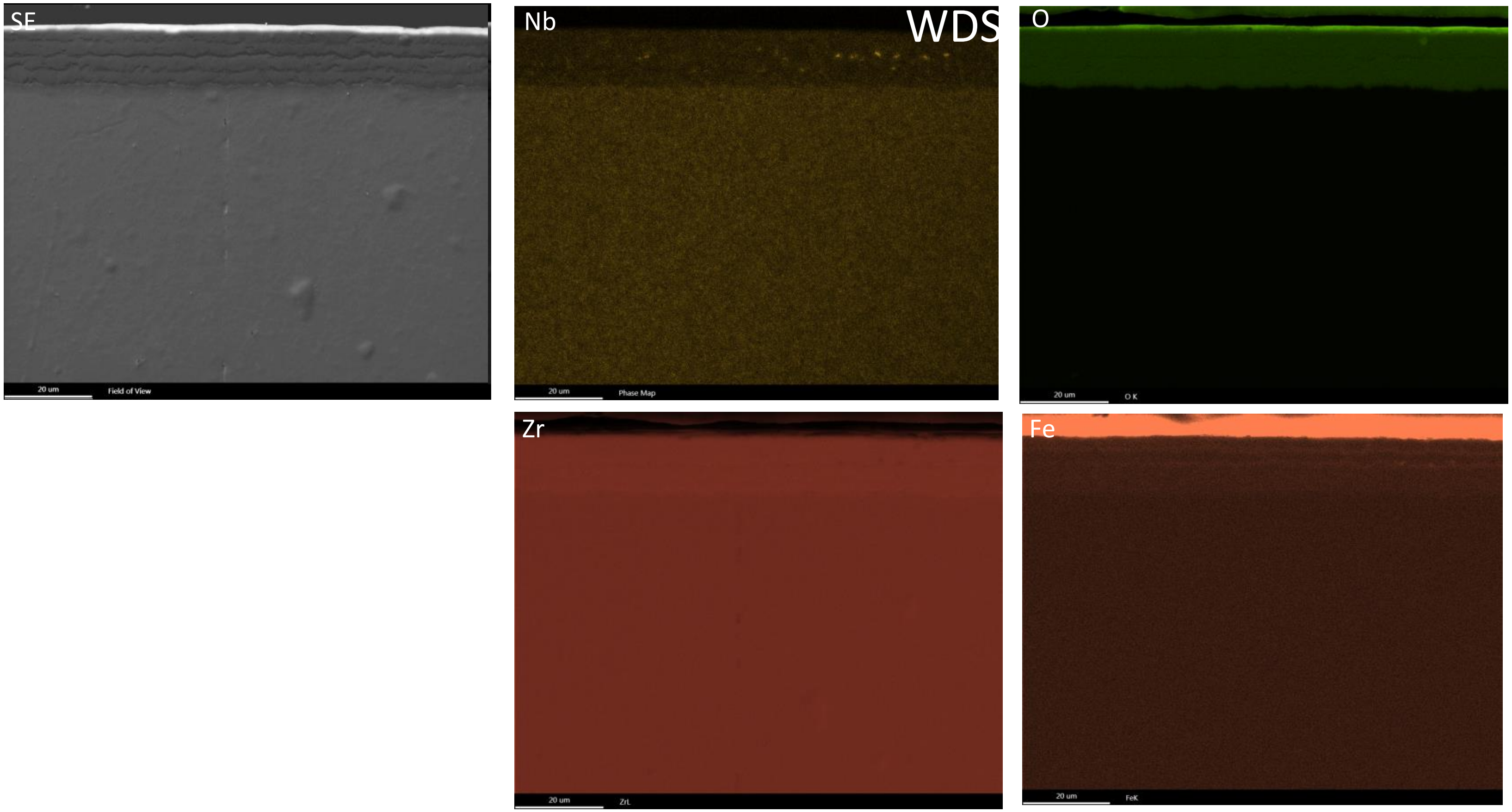
- Mottled appearance in Nb map indicated the presence of SPPs but too small to be detected individually using SEM/EDS/WDS

SAMPLE E110OPT – NB & ”LOW” FE – E/WDS MAPPING



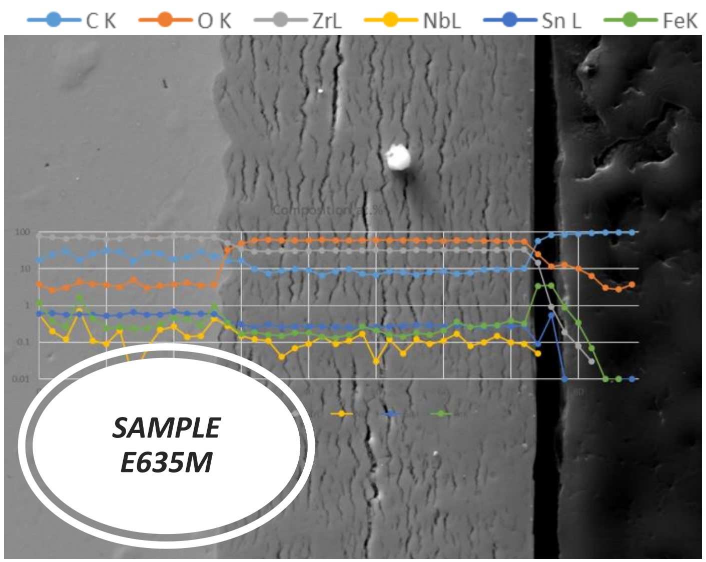
- No SPPs or other variations in elemental composition could be observed

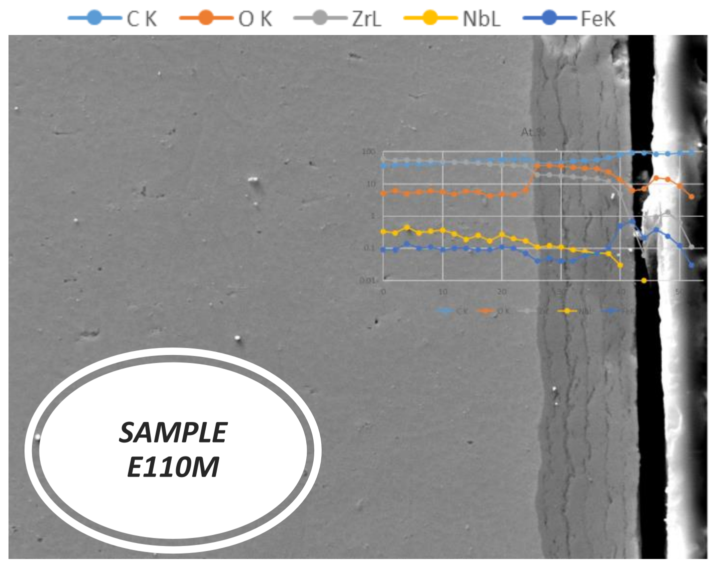
SAMPLE E125 – HIGH NB AND ”LOW” FE – E/WDS MAPPING

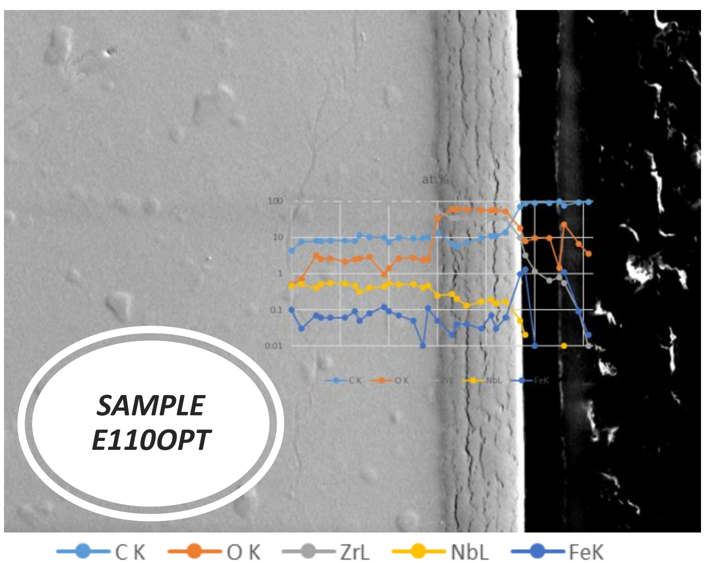


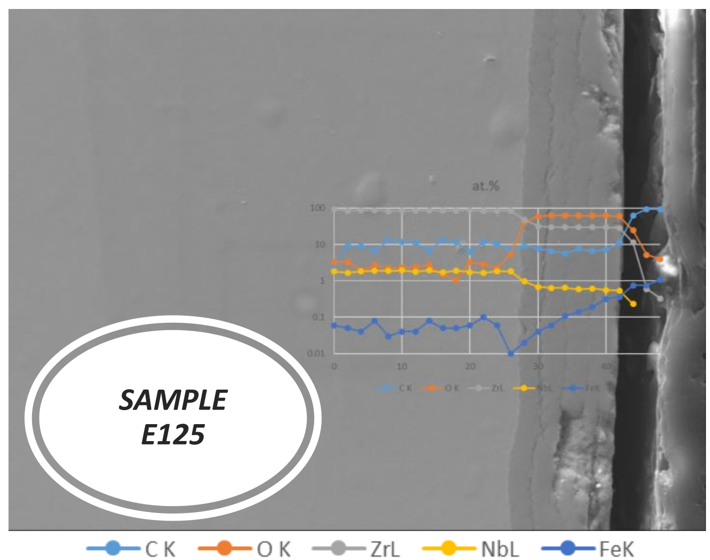
- Nb (and Fe) enrichment in the oxide layer – possible precipitation in oxide?

OXIDE QUANTITATIVE ELEMENTAL COMPOSITION









- Oxides homogenous regarding elemental composition in agreement with expected values
- No impurities or contaminants observed
- Possibly the Fe content increase towards the outer part of the oxide – observed for E110M, E110Opt and E125, not as obvious for E635M

TEM ANALYSES

- A JEOL 2100F equipped with an Oxford EDS and Gatan EELS are used for the analyses.

- 3 mm in diameter TEM discs were prepared from the metal part of the alloys using standard grinding, polishing, punching and electrolytic polishing techniques. Lamellas covering the complete oxide thickness was prepared using the focused ion beam (not covered in this presentation)

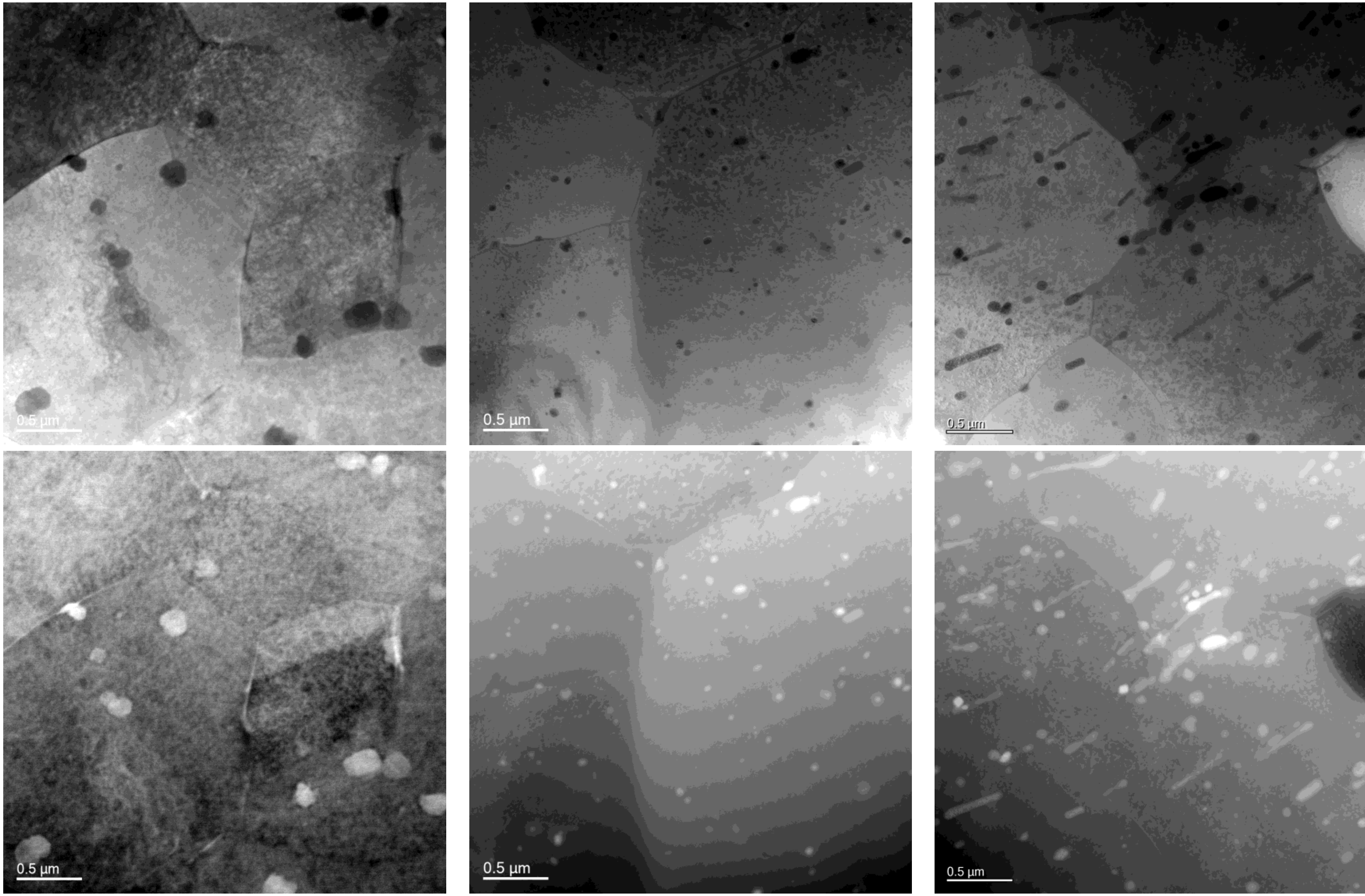
- Only initial data will be presented – TEM analyses on-going.

- Focusing on SPP chemistry and density, <a>- and <c>-type dislocations.

SPP DISTRIBUTION

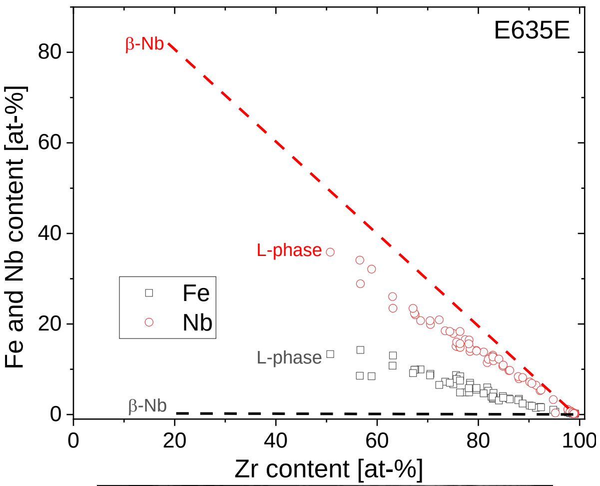
BF

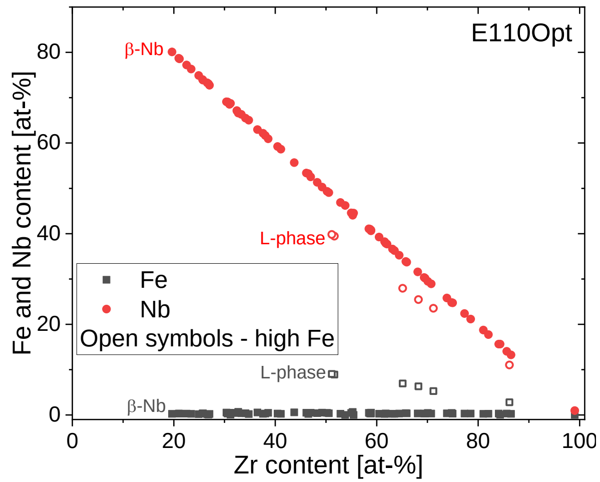
DF

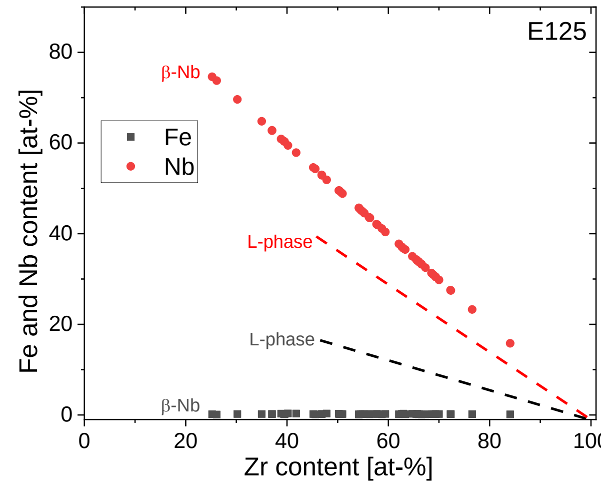


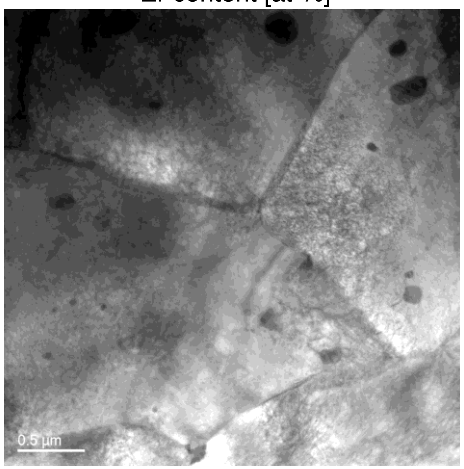
Sample E635M | Sample E110Opt | Sample E125

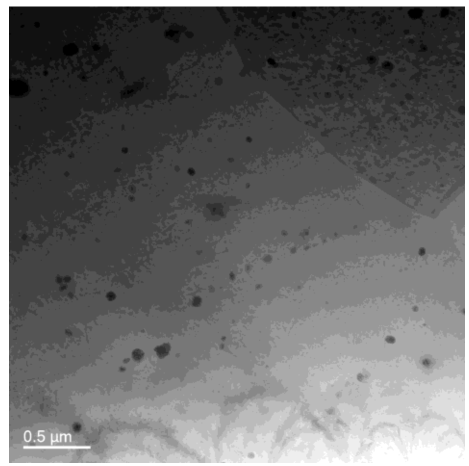
SPP ELEMENTAL COMPOSITION

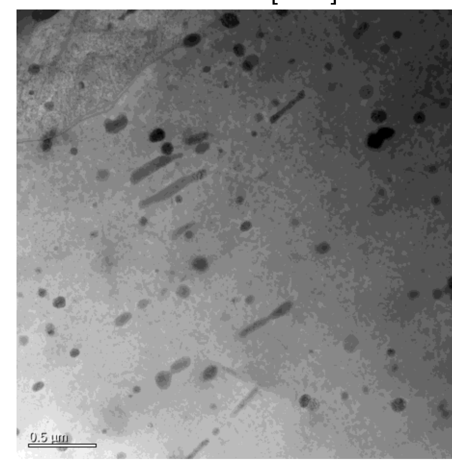






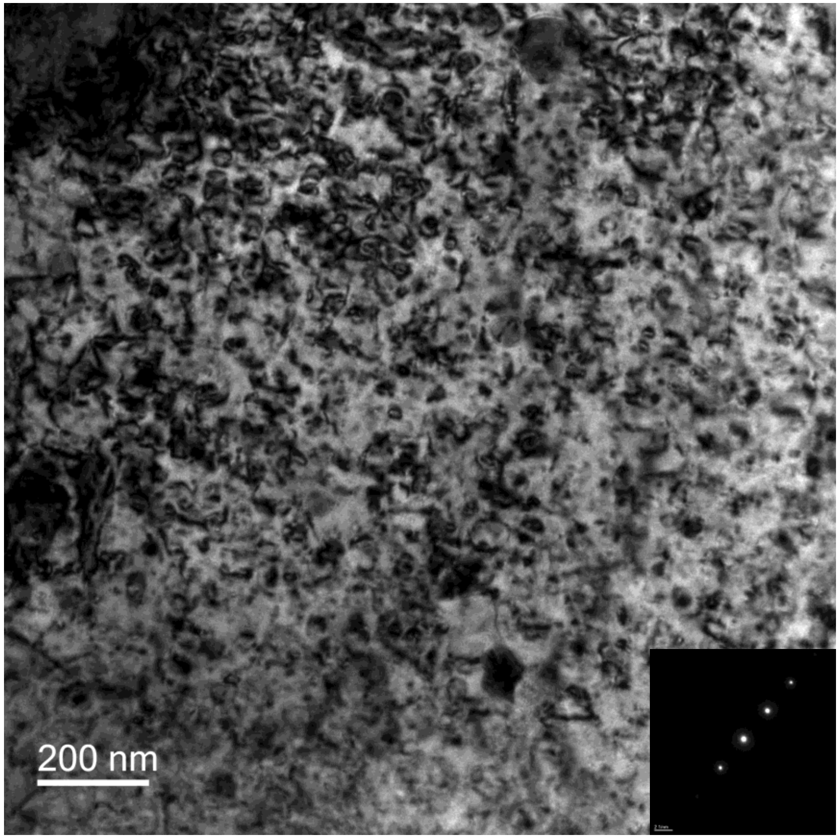


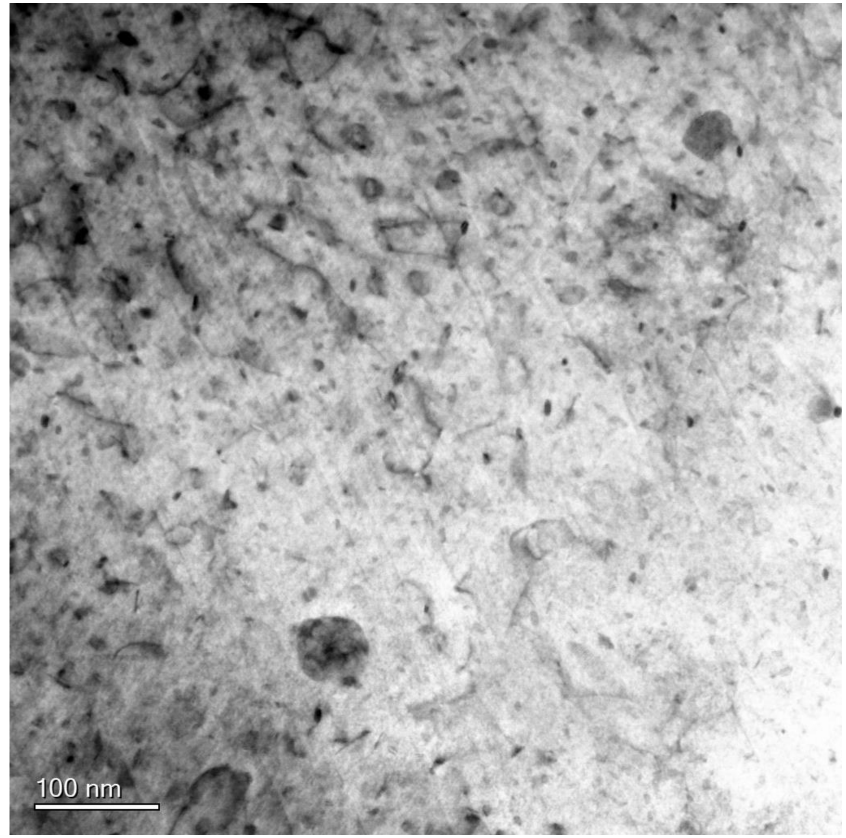




- The three alloys have a different set of SPPs after irradiation
- Supports the fact that the Laves phase particles dissolve during irradiation

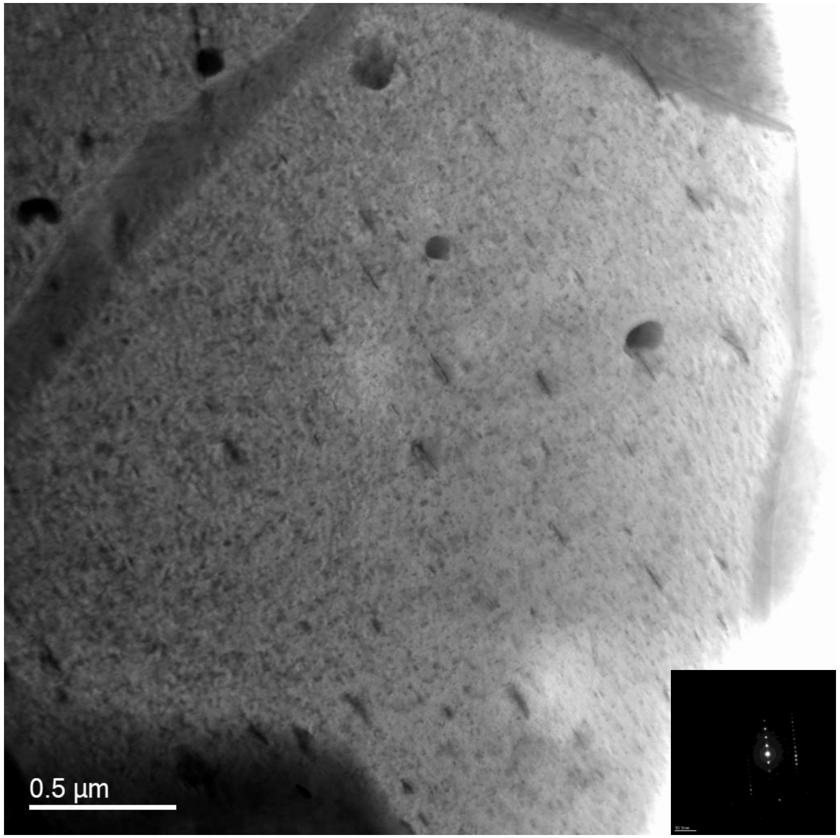
IRRADIATION INDUCED DISLOCATION <A>-LOOPS

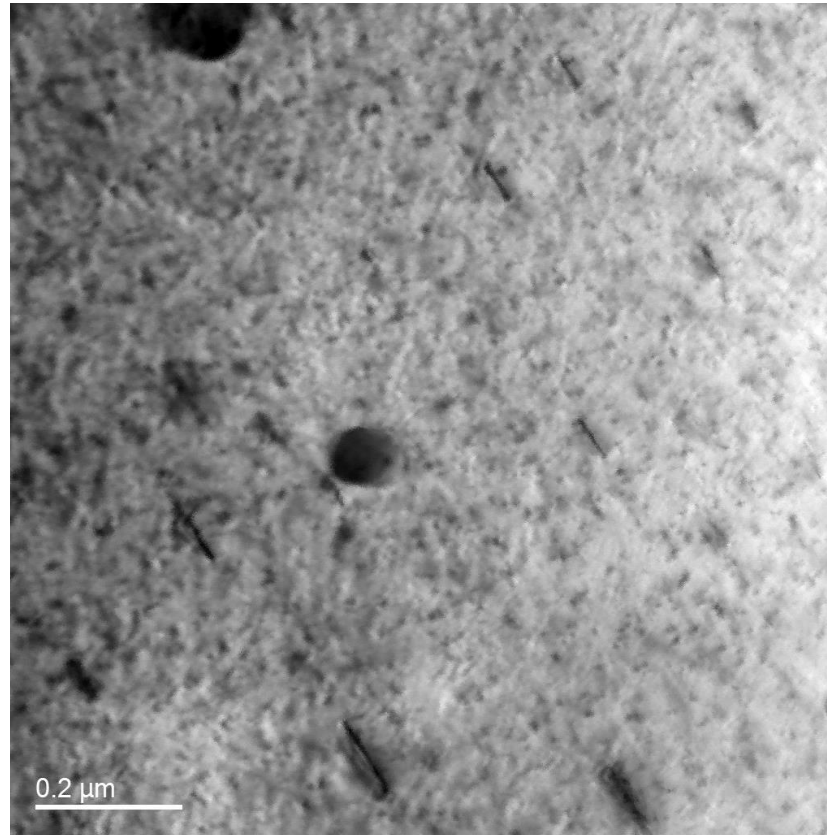




- A very high density of <a>-component loops present in all material
- Still under evaluation regarding density and comparison between different alloys.

IRRADIATION INDUCED DISLOCATION <C>-LOOPS





- A low density of <c>-component loops present. Possibly the slightly higher than normal irradiation temperature affects the onset of <c>-loop formation…?
- Still under evaluation regarding density and comparison between different alloys.

SUMMARY

- SEM shows that alloy E635M corrodes the most in reactor
- The oxide morphology is different depending on alloy composition. More small pores are present in the high Nb alloy E125.
- Dissolution of particles seems to take place in the oxide layer. The oxide layer for Alloy E635M show clear SPPs in the inner of the oxide but none in the outer parts
- Nb rich precipitates have formed in the high Nb alloy E125 oxide layer
- Fe levels higher in the outer part of the oxide layers indicate possible uptake from external sources.

FUTURE WORK

- Continue TEM examinations on the base Zr-based alloy
- Perform the analyses on the oxide samples
- Continue the Laser Ablation experimental work, assess impact of lithium uptake on alloy corrosion
- Further evaluation of the data

THANK YOU FOR YOUR ATTENTION!