INL/MIS-24-78202-Revision-0

Characterization of Legacy Fuels: Progress to Fuel Cladding Chemical Interaction

July 2024

Luca Capriotti, Kyle Paaren, Daniele Salvato, Fidelma Giulia Di Lemma, Yachun Wang, Fei Xu, Tiankai Yao, Douglas L Porter

Changing the World's Energy Future

Idaho National Laboratory

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Characterization of Legacy Fuels: Progress to Fuel Cladding Chemical Interaction

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Idaho National Laboratory

*now at Lightbridge Corporation

Metal Fuel workshop, Idaho Falls, May 14-16th

U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Metallic Fuel R&D Priorities – FCCI Introduction

Technical R&D Gaps on the Reference Design: Improving FCCI understanding and database reliability

- **FCCI phenomenon introduction and state of the knowledge:**
  - It results from interaction between fuel elements (U, Zr, Pu), fission products (lanthanides, Ln), and cladding elements (mostly Fe in HT-9)
  - Thermodynamically & Irradiation assisted: temperature, burnup, power, time
  - Formation of eutectic interaction products (Fe-U) → lower melting point
  - Formation of brittle layers (Ln-Cladding) → reduce cladding strength → Cladding Wastage
  - Several papers and reports reviewed the knowledge base and PIE data*
    - Phenomenon empirically understood fairly well

➢Identified as life-limiting phenomenon and key fuel performance in design basis efforts**

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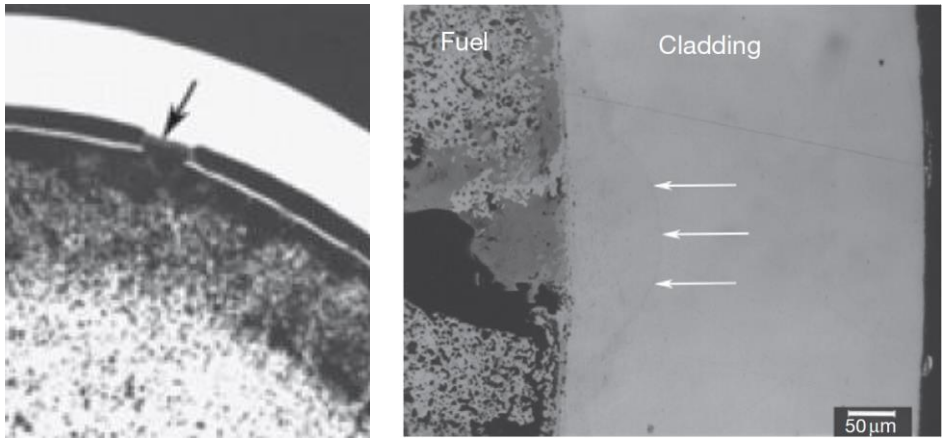
*Keiser, Metal fuel-cladding interaction, Comprehensive Nuclear Materials, 2012 / 2020
*Keiser, Fuel cladding chemical interaction in metallic sodium fast reactor fuels: A historical perspective, JNM, 2019
*Matthews, Fuel-Cladding Chemical Interaction in U-Pu-Zr Metallic Fuels: A Critical Review, Nuc Tech, 2017
**NUREG/CR-7305;
**Porter & Crawford, Fuel Performance Design Basis for the Versatile Test Reactor, Nuc Sci and Eng., 2022

FCCI knowledge gaps

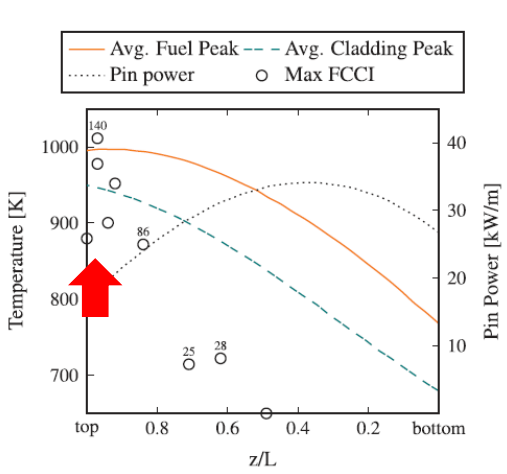
- **FCCI characterization from literature data:**
  - Mostly optical microscopy based with uncertainties in phase contrast and discrimination.
  - Localized analysis and limited spatial resolution;
  - No chemical/crystallographic information;
  - Missing statistical details (max, average) across entire interface
  - Limited data at different irradiation conditions (T, burnup, power)
  - Very limited data available for prototypic fuel pin design (e.g. MFF/IFR-1) vs EBR-II fuel design. → FCCI peak vs T, BU



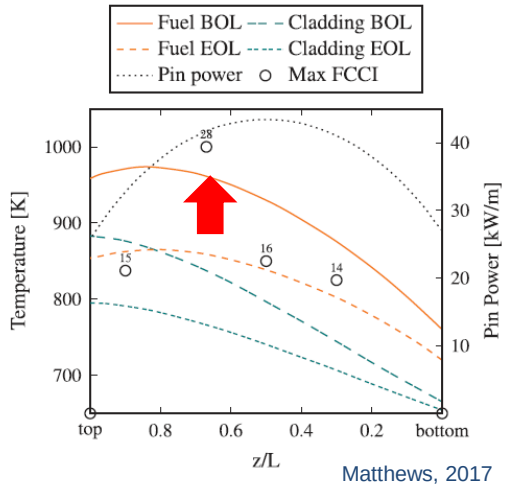
Examples of optical images on FCCI regions

Overall, data not well documented and scarce leading to high uncertainties in design equations.

EBR-II X447 DP-70



FFTF IFR-1



Matthews, 2017

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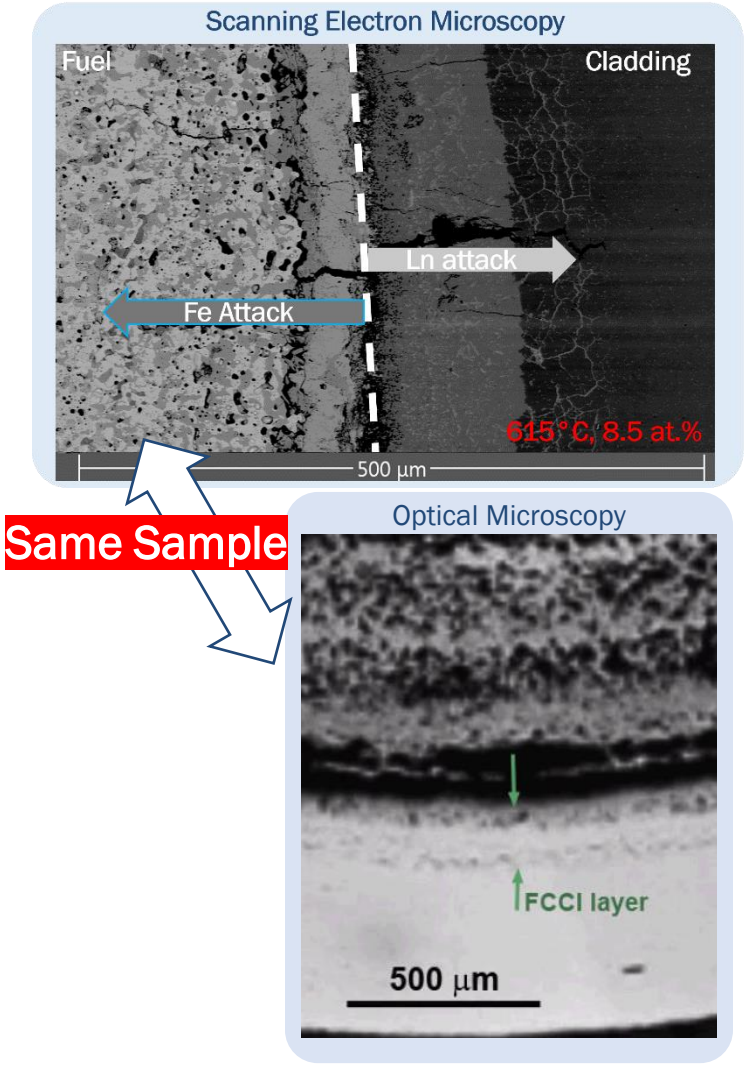
FCCI R&D and Objectives

- **FCCI R&D:**
  - Electron microscopy techniques (e.g. SEM) found to provide clear contrast for FCCI zones
  - Prototypic length FFTF pins (IFR/MFF experiments) higher priority to harvest
  - BISON simulations providing evaluation of MFF pin performance to compare with PIE results and guide PIE exams
  - Improve understanding on chemical and mechanical performance of the various layers of FCCI.
  - Utilization of new methodology for data analysis.

- **Objectives:**
  - Achieve a reliable, reproducible, statistically-robust analysis of FCCI depth and characteristics under various irradiation conditions
  - Reduce uncertainties in design equations for fuel performance code
  - Improve predicting capabilities of fuel performance code modeling FCCI



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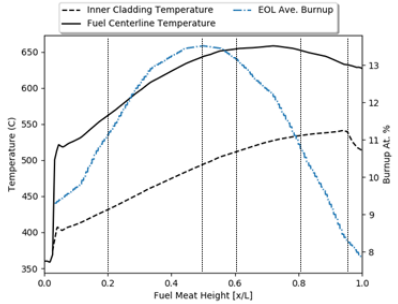
A holistic approach

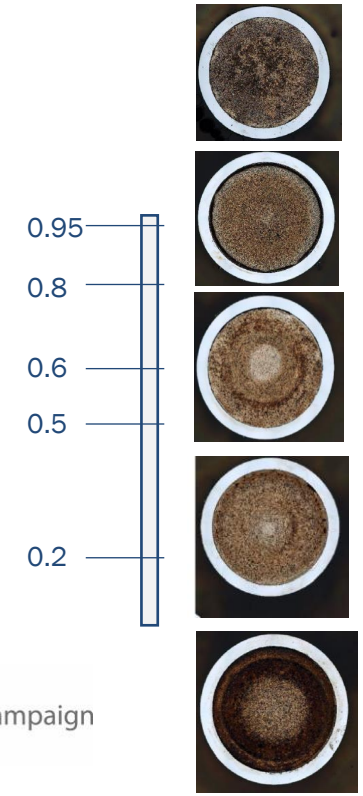
MFF Irradiation Test

BISON Simulations

T, burnup

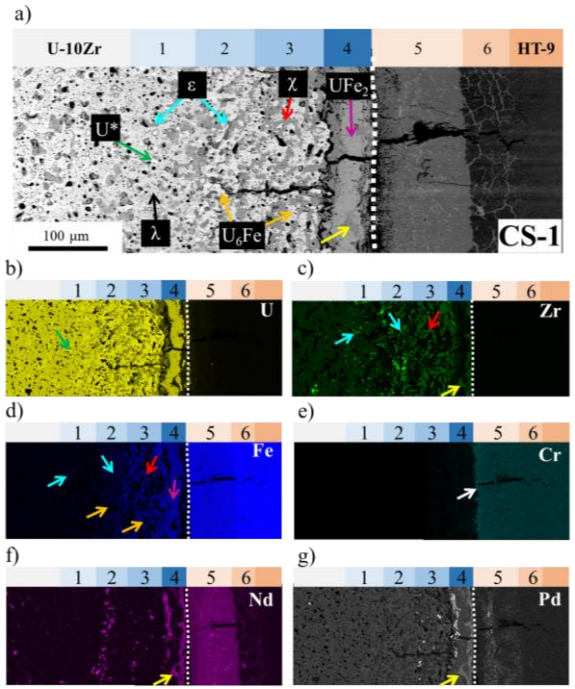


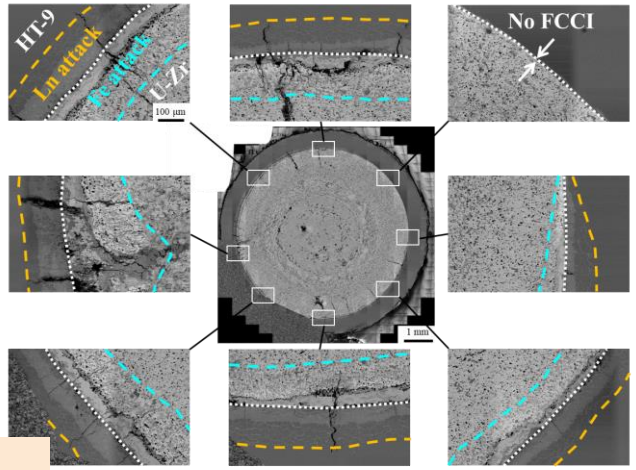
Samples Selection



Pin Cutting and Cross-Section Preparation

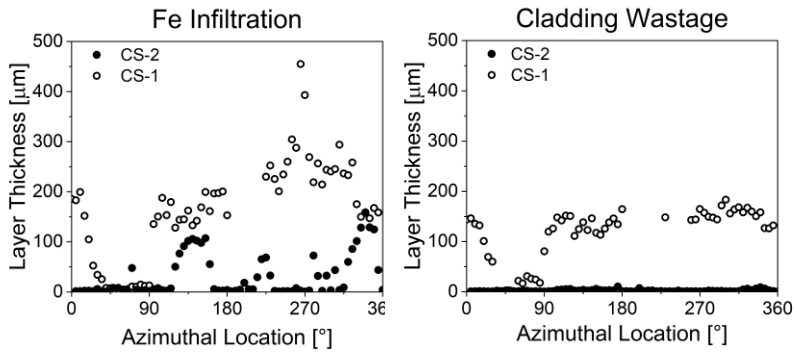
SEM – BSE/EDS
& mech. testing





Computer-assisted
analysis of PIE Data

FCCI Characterization



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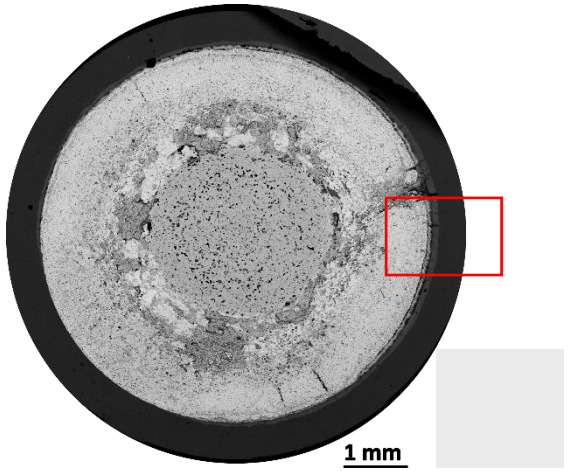
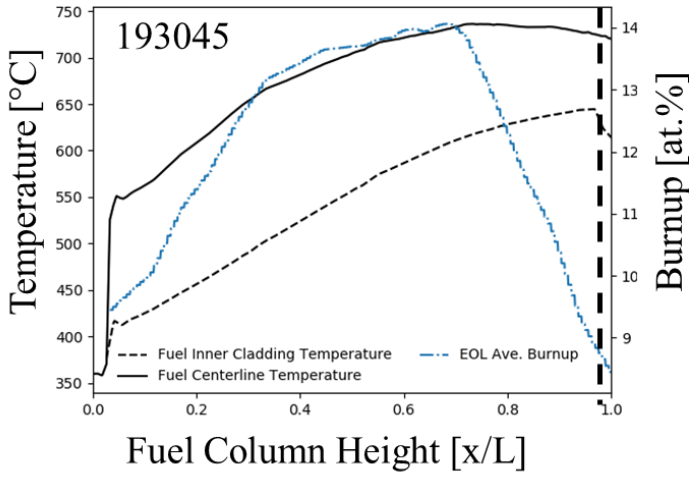
FCCI vs Temp and BU: MFF3 - high Temp, low BU

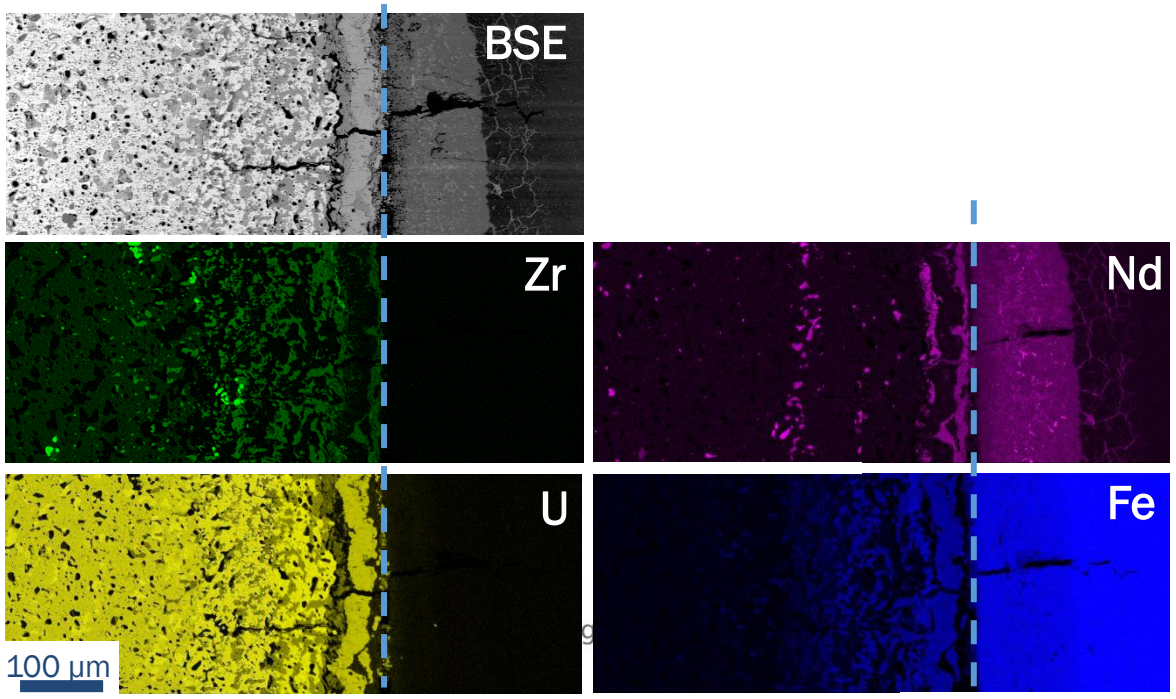
Salvato et. al, The effect of temperature and burnup on U-10Zr metallic fuel chemical interaction with HT-9: A SEM-EDS study, JNM, 2024

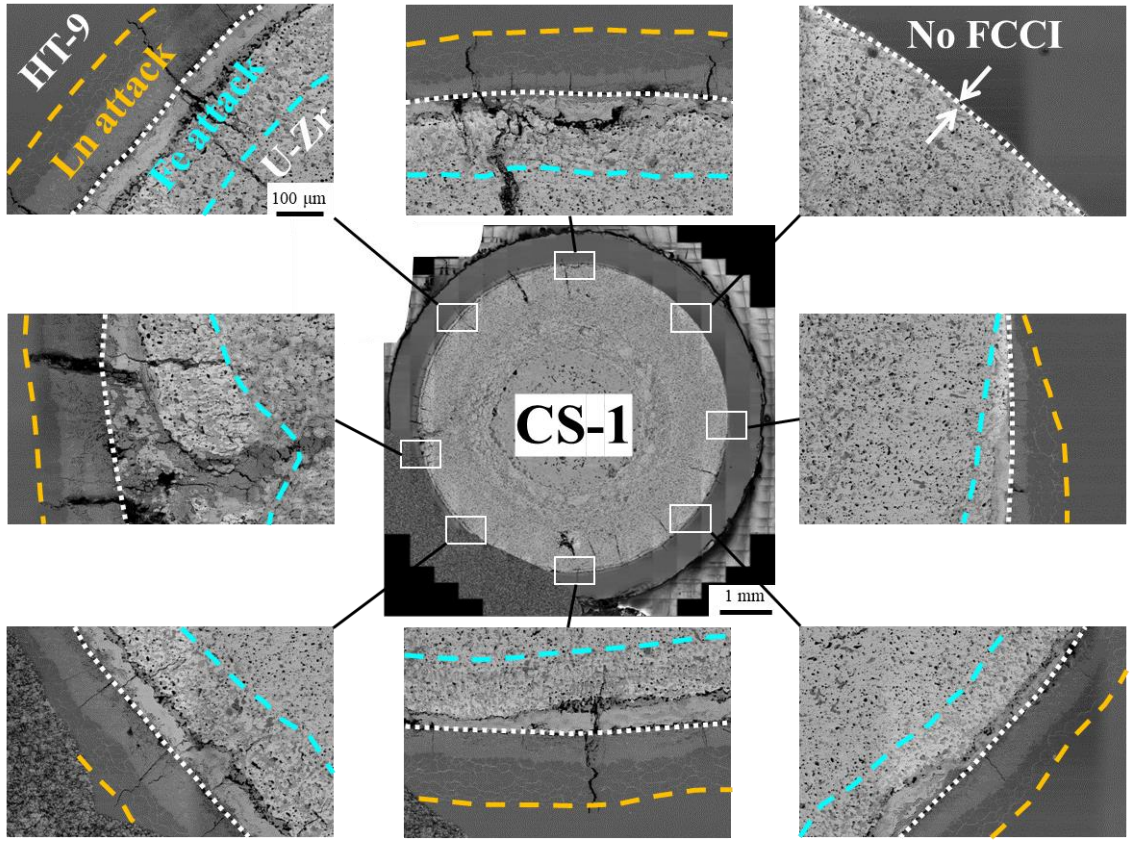




MFF3 193045, cross section 1 (CS1), x/L=0.98, 8.5% FIMA, clad T 615 °C:

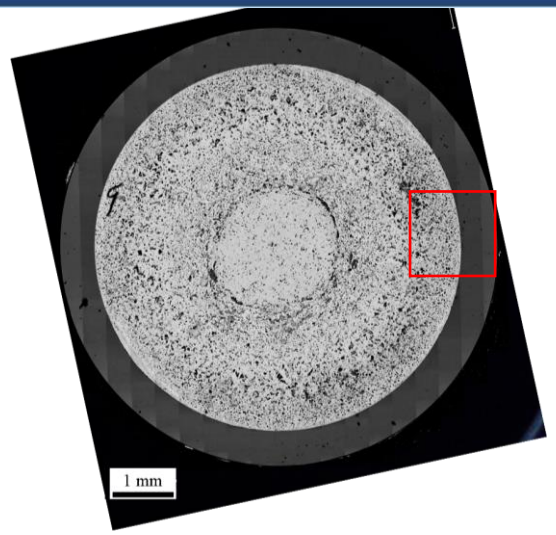
- Intense Fe infiltration into the fuel.
- High Ln infiltration into the cladding: uniform layer + intragranular wastage with Ln (due to coarsened grains from carbon loss)

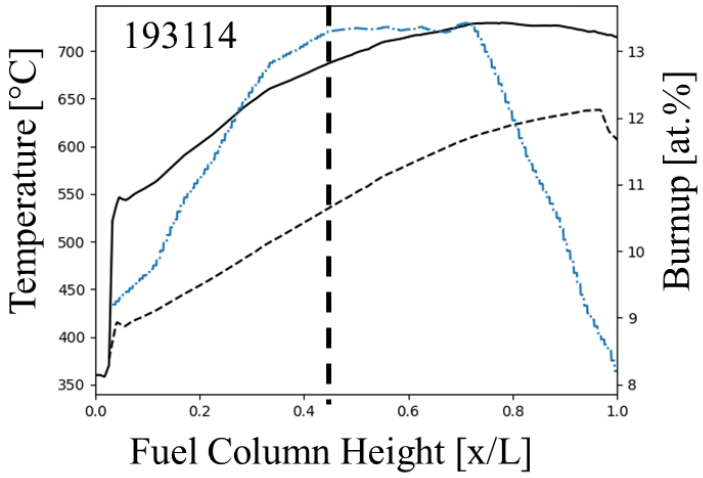




FCCI vs Temp and BU: MFF3 - low Temp, high BU

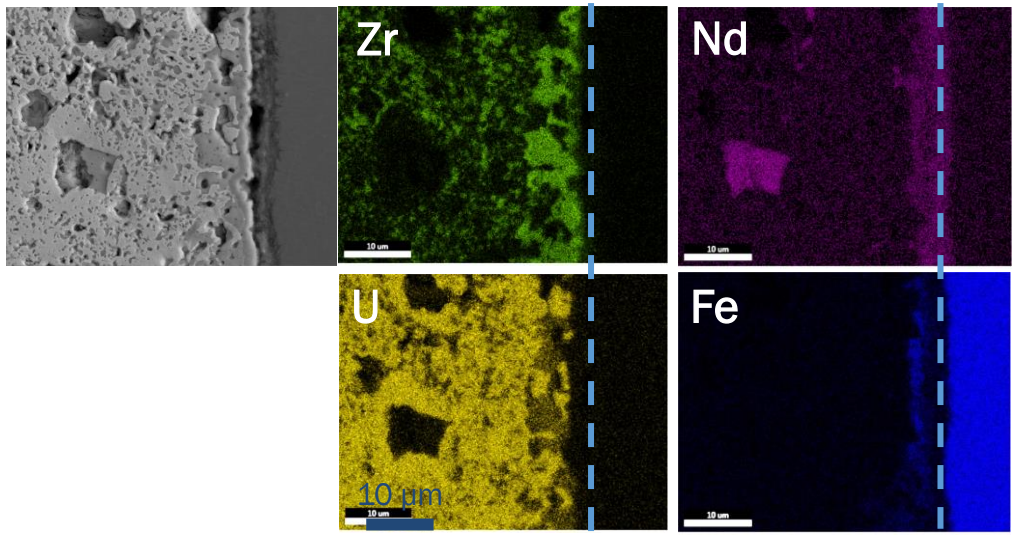
Salvato et. al, The effect of temperature and burnup on U-10Zr metallic fuel chemical interaction with HT-9: A SEM-EDS study, JNM, 2024

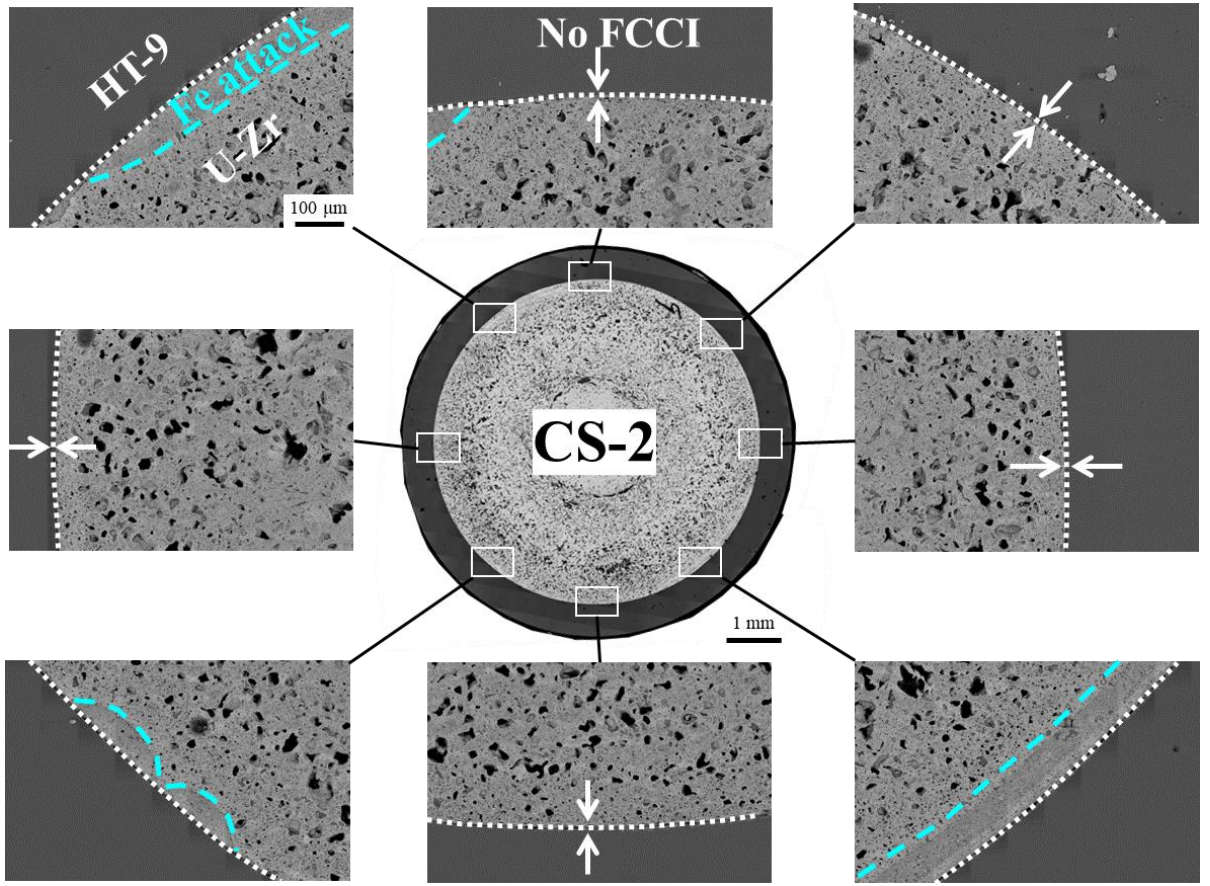




MFF3 193114, cross section 2 (CS2), x/L=0.44, 13.2% FIMA, Clad T 530 °C:

- Limited (<10 μm) lanthanides (Ln) attack (cladding wastage) throughout all interface;
- Localized regions of deep (100-150 μm) Fe infiltration into fuel leading to layered-microstructure





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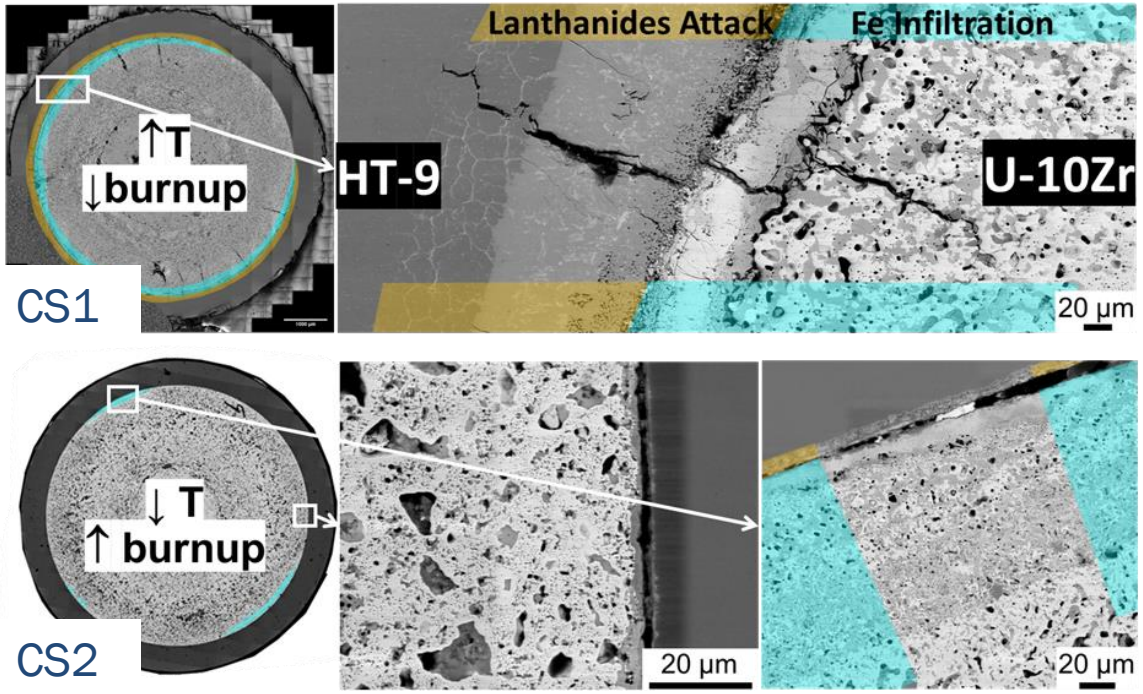
FCCI vs Temp and BU: MFF3 comparison

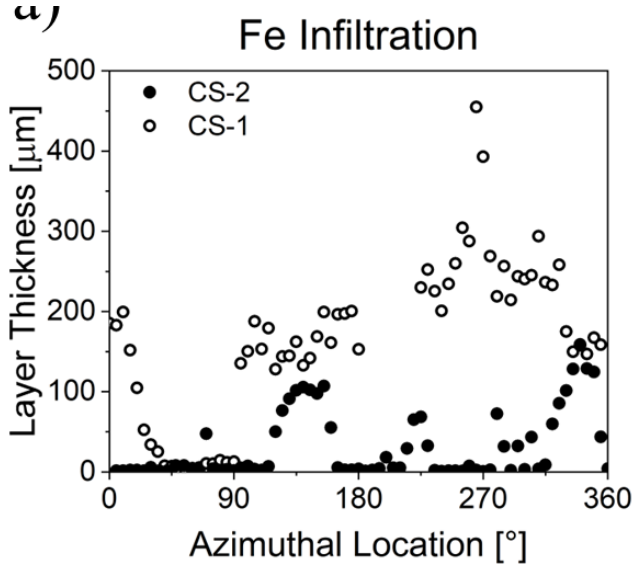
Salvato et. al, The effect of temperature and burnup on U-10Zr metallic fuel chemical interaction with HT-9: A SEM-EDS study, JNM, 2024

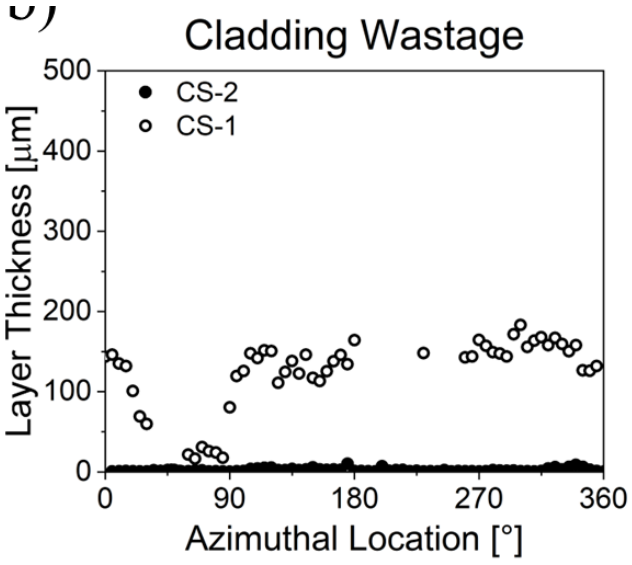


FCCI vs Temp and BU – MFF3 comparison summary

- Manually measured wastage every 5 degree.
- Estimated Fe infiltration from EDS maps / line scans
- Striking different between the 2 CS – still data missing from 0.6-0.9 x/L to confirm FCCI peak
- Data is being used to V&V empirical and (new) mechanistic code in BISON - MARMOT

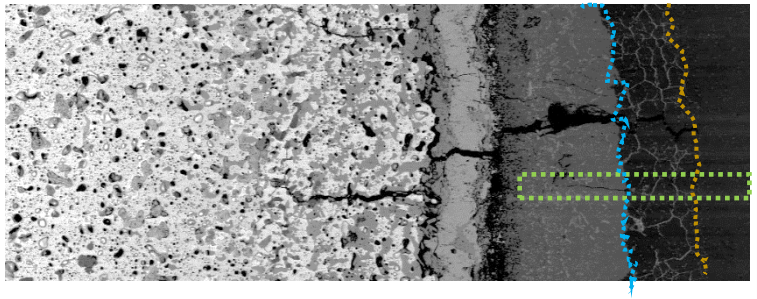
		CS-1	CS-2
Fe Infiltration	Average (μm)	164	31
	SD (μm)	98	42
	Max value (μm)	455	159
Cladding Wastage	Average (μm)	115	2
	SD (μm)	54	2
	Max value (μm)	183	10

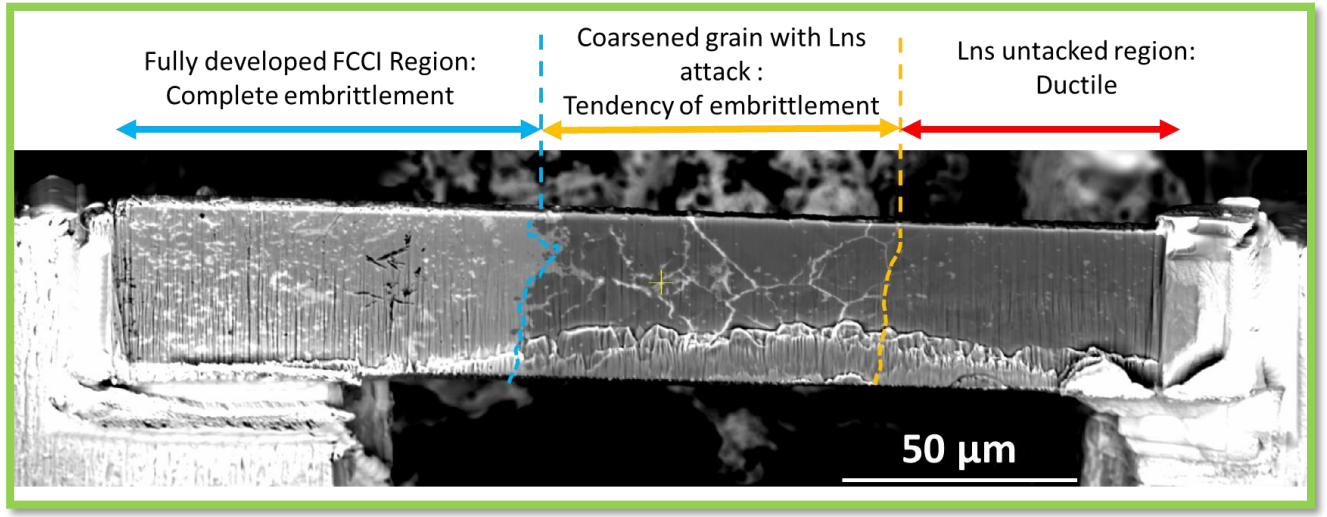


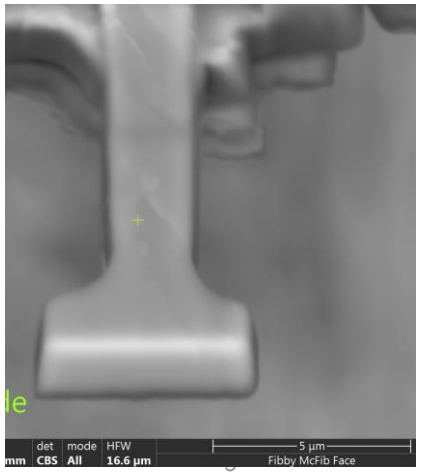


FCCI - mechanical properties of wastage layers

- Small-scale mechanical testing is capable to probe the location-specific mechanical properties of materials of limited volume, like the FCCI zone.
- Micro tensile testing revealed the mechanical property changes in the inner lift-out, while very consistent behavior was observed for tensile specimens in the outer lift-out (HT9 not affected, not shown here)
- For MFF3 CS1 radial cross section sample results (see also central image below):
  - Embrittlement in fully developed FCCI layer
  - Tendency of embrittlement in coarsened grain (due to carbon loss) with GBs attacked by Ln
  - Mechanical softening cross the HT-9 matrix
- Post-testing TEM characterization revealed a very complex microstructure and phases formation
  - Example, in fully developed FCCI layer, 3 intermetallic phases formed, Fe2Ce, sigma_FeCr, and a ternary Fe8.84Cr2.60Ce1.56 phase, causing embrittlement.







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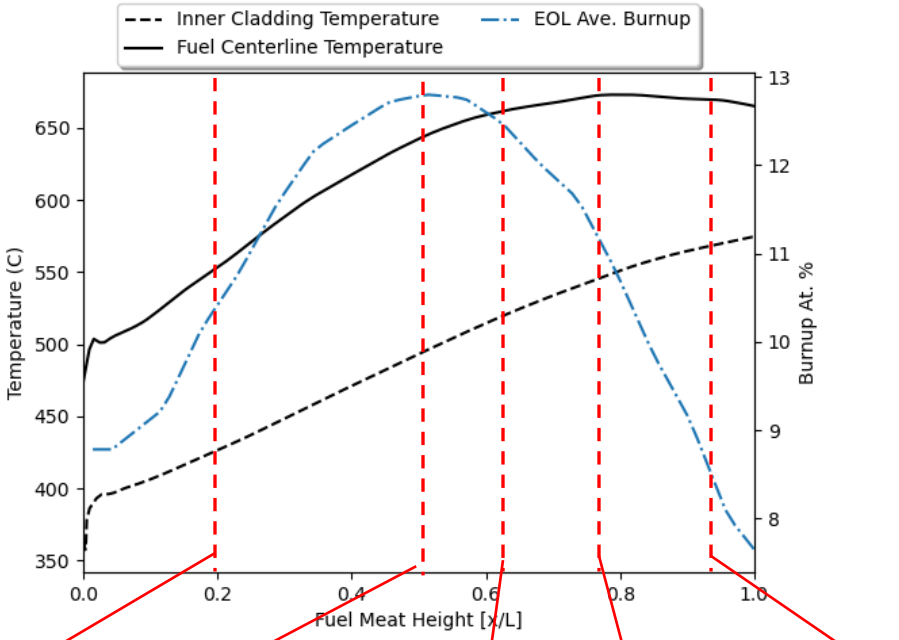
FCCI – Confirming low temp behavior



0.20 x/L | 0.50 | 0.63 | 0.78 | 0.95

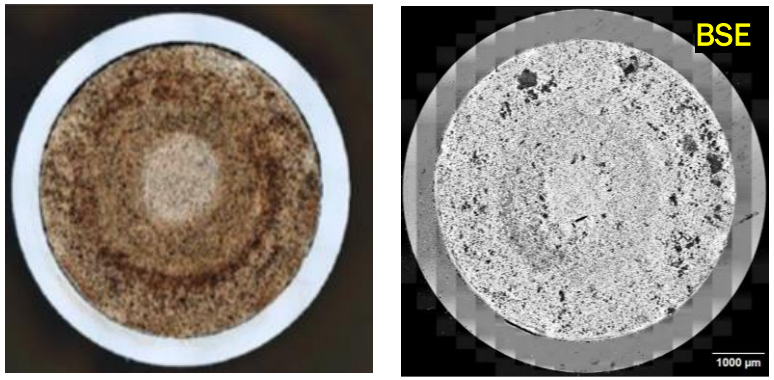
T_{ic} ~ 495 °C

T_{ic} ~ 520 °C

MFF2 192167:

- 5 sections of interest identified with modelling support
- To confirm: low or not-existing wastage level at these temp regime
- 2 cross section already examined via SEM
- Other cross sections in progress

0.63 x/L



0.50 x/L

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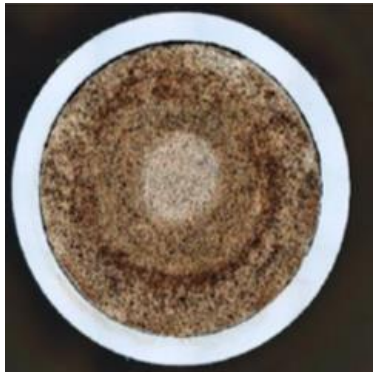
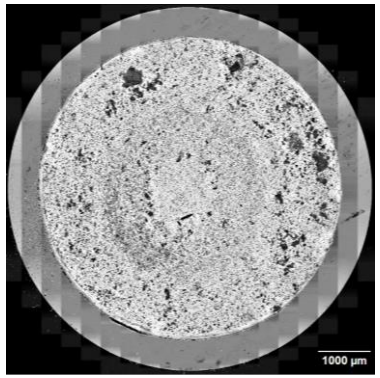
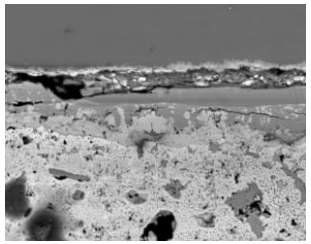
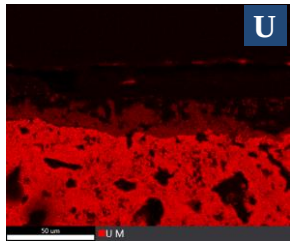
FCCI – Confirming low temp behavior

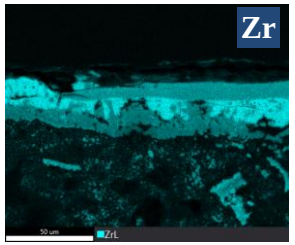
0.63 x/L

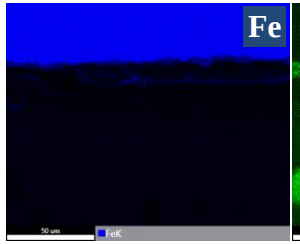


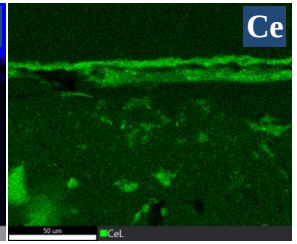




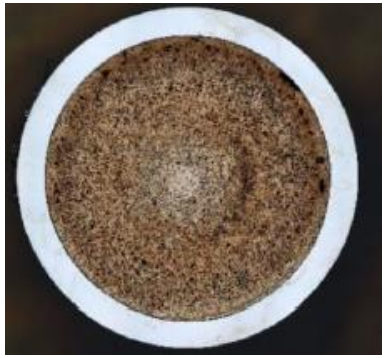


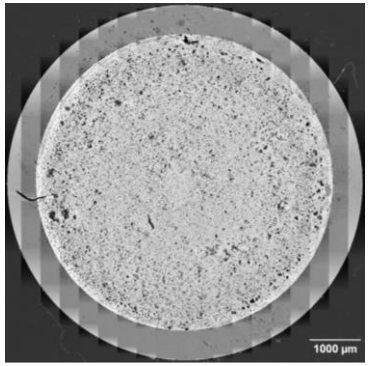


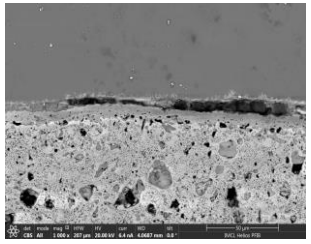


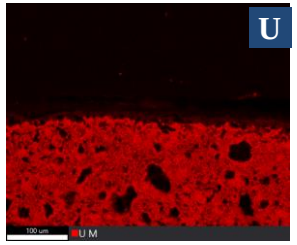


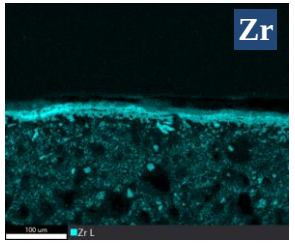
0.50 x/L

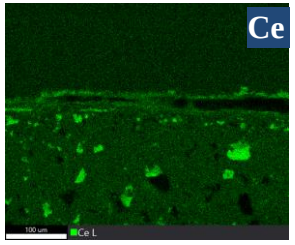












MFF2 192167, 2 cross section analyzed so far:

- Minimal lanthanides (Ln) attack (cladding wastage) throughout all interface;
- Limited Fe infiltration into fuel
- 0.50 x/L cross section present almost no Zr-redistribution
- Wastage layer/s quantification in progress

Computer-assisted analysis of FCCI wastage

- Developed a new artificial intelligence (AI) model to assist manually annotating and detecting the interesting objects on multi-scale and multiple instrumental microscopies*. The flowchart of the interactive AI model based on natural images is shown in Fig. 1
- The model was applied to assist the FCCI thickness analysis.
  - The two wastage layers were detected accurately with several clicks (needs less 5 minutes) by experts as shown in Fig. 2.
    [from CS1: layer 5= fully developed FCCI; layer 6= coarsened grain with Ln]

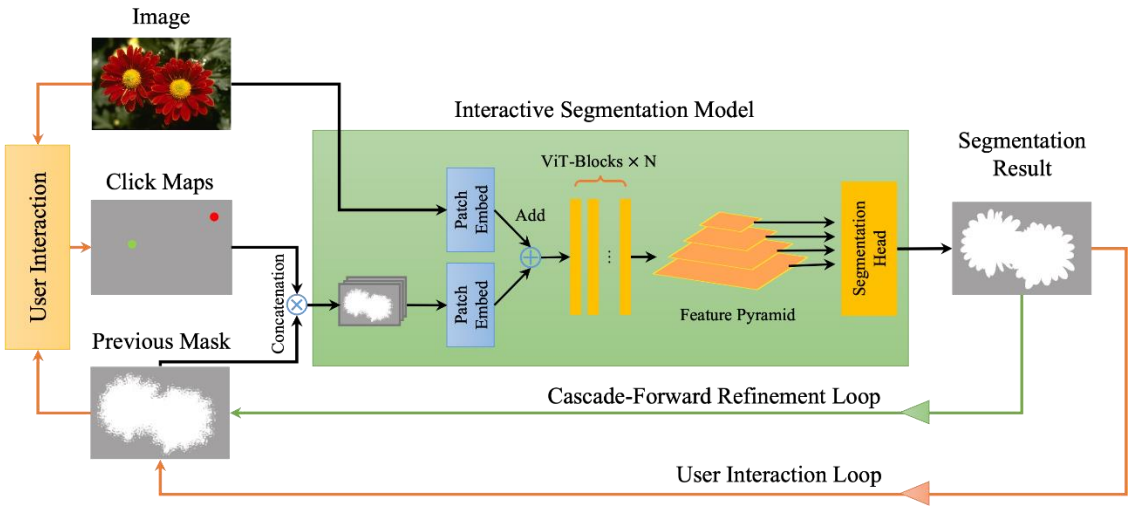


Fig. 1 Overview of the model. In the click map, the green color click is in the target object; the red color click is from background.

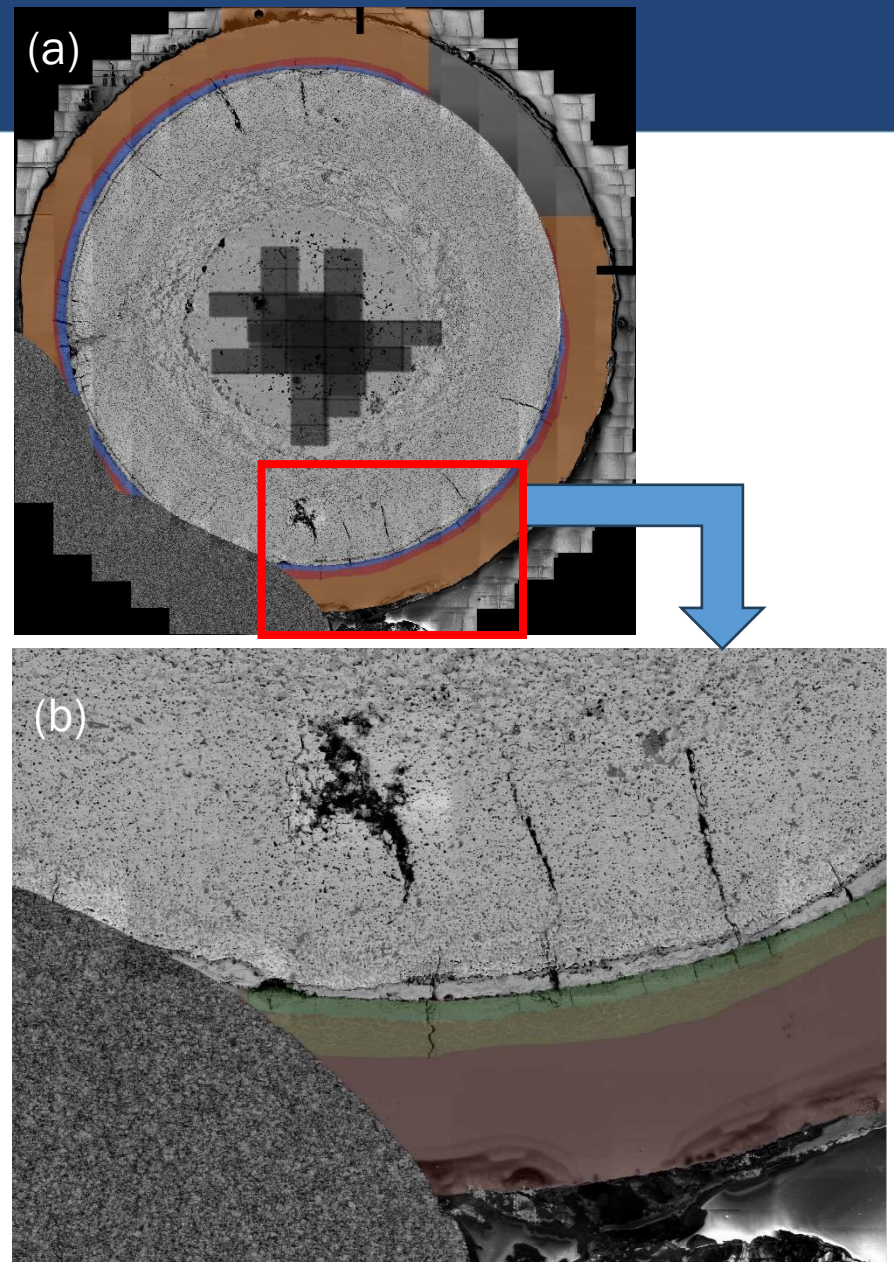


Fig. 2. wastage layers detected by the model. (a) shows the detection result of the cross-section. (b) one zoomed in image patch result. Red color shows the cladding area, green color illustrates the layer 5; and the yellow color shows the layer 6.

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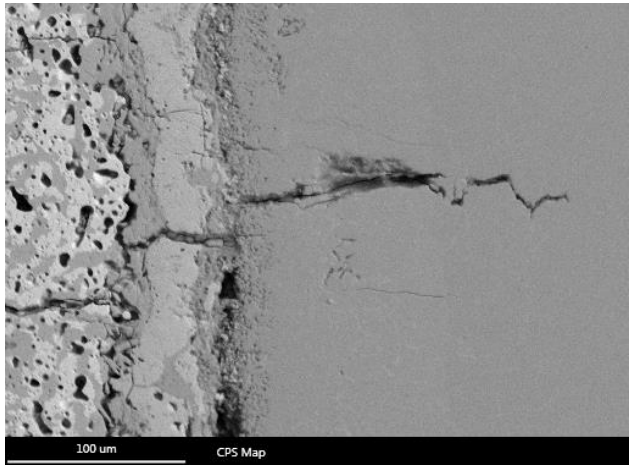
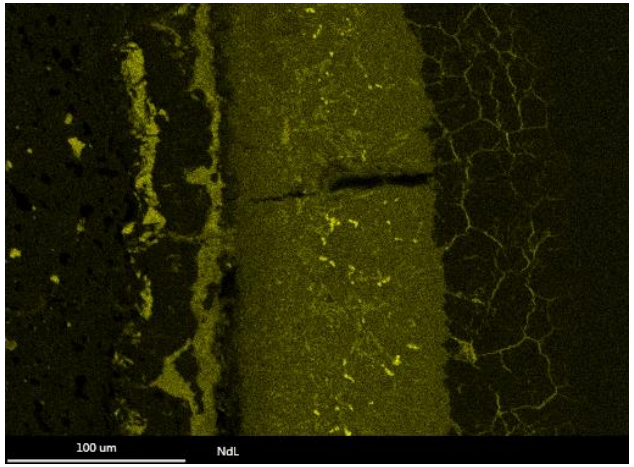
*S Sun, M Xian, F Xu, T Yao, L Capriotti. CFR-ICL: Cascade-Forward Refinement with Iterative Click Loss for Interactive Image Segmentation. AAAI 2024: 5017-5024; 2023.

Comparisons of FCCI wastage layer detection

BSE image



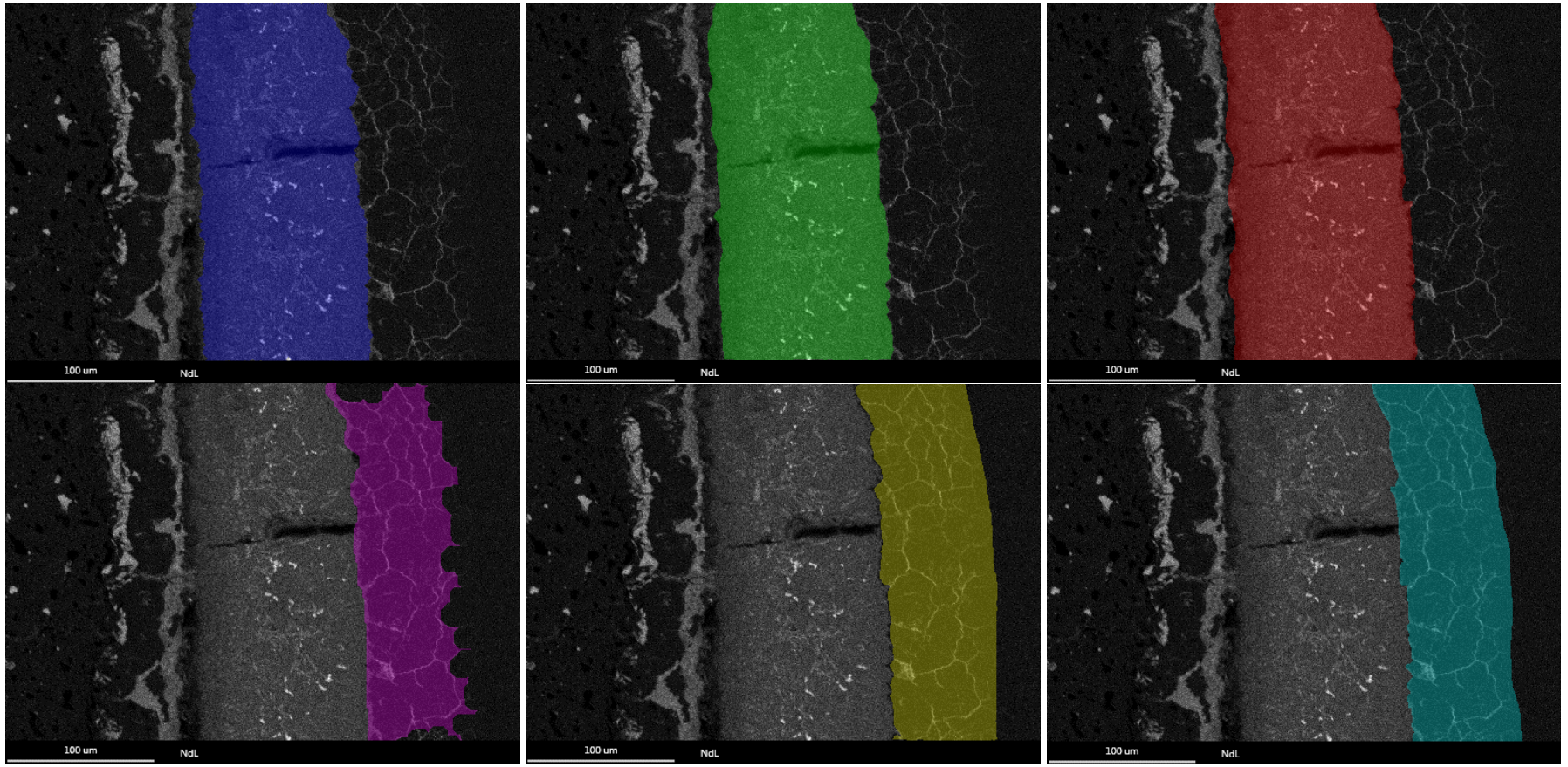
Nd EDS map



Detected from EDS

AI model results

Expert manually labeled



First row shows the detection results from different methods on layer 5; second layer was the comparison of layer 6. The visualization findings are that the detection of layer 5 was consist with three different ways, but layer 6 was not.

Computer-assisted analysis of FCCI wastage – path forward

- Using the AI model to label the FCCI layers is more efficient comparing with manual labeling from scratch.
- The layers generated from the model are more objective after given confident several inputs from experts
- On-going evaluation of the model for prediction vs “real” / actual results
- The labelled data could be utilized to train deep learning models which will generate layer 5 and layer 6 results by given any BSE images with cladding without use of user labelling.

Preliminary results comparing the computer-assisted method vs EDS image process and expert manual

Methods	layer	Minimum(μm)	Maximum(μm)	Average (μm)	Standard deviation(μm)
Detection on EDS using image processing method	Layer 5	83.56	113.42	105.43	5.87
	Layer 6	34.29	50.19	40.89	3.18
AI model	Layer 5	91.78	114.98	108.53	4.57
	Layer 6	65.92	77.39	72.16	2.59
Manual labeled	Layer 5	103.74	122.63	114.85	4.17
	Layer 6	57.74	71.31	65.40	3.41

Conclusion

- Technical R&D Gaps on the Reference Design: Improving FCCI understanding and database reliability
- Identified as life-limiting phenomenon and key fuel performance in design basis efforts
- Historical data not well documented or missing for prototypical fuel design (e.g. FFTF MFF)

- Obj are
  - Achieve a reliable, reproducible, statistically-robust analysis of FCCI depth and characteristics under various irradiation conditions
  - Reduce uncertainties in design equations for fuel performance code
  - Improve predicting capabilities of fuel performance code modeling FCCI

- Holistic approach with BISON informing selection on harvesting legacy materials
  - Using SEM for FCCI wastage characterization, quantification
  - Harvesting FFTF MFF materials
  - Using computer-assisted analysis on FCCI wastage
- FCCI vs Temp, BU: full formation of Ln and intragranular wastage (@high temp, low BU)
- FCCI – low temp wastage characterization: limited or not-existing interaction.
- Computer-assisted wastage analysis: AI model developed and compared with expert manual annotation and EDS. Model evaluation in progress.

Questions?

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