Advanced 3D Characterization and Reconstruction of Reactor Materials

Assel Aitkaliyeva[1], Benjamin Hauch[2], Bradley Fromm[1,3], Kumar Sridharan[2]

1. Idaho National Laboratory
2. University of Wisconsin
3. Washington State University

Contact:
E-mail: assel.aitkaliyeva@inl.gov
Tel: 208-533-8853

www.inl.gov

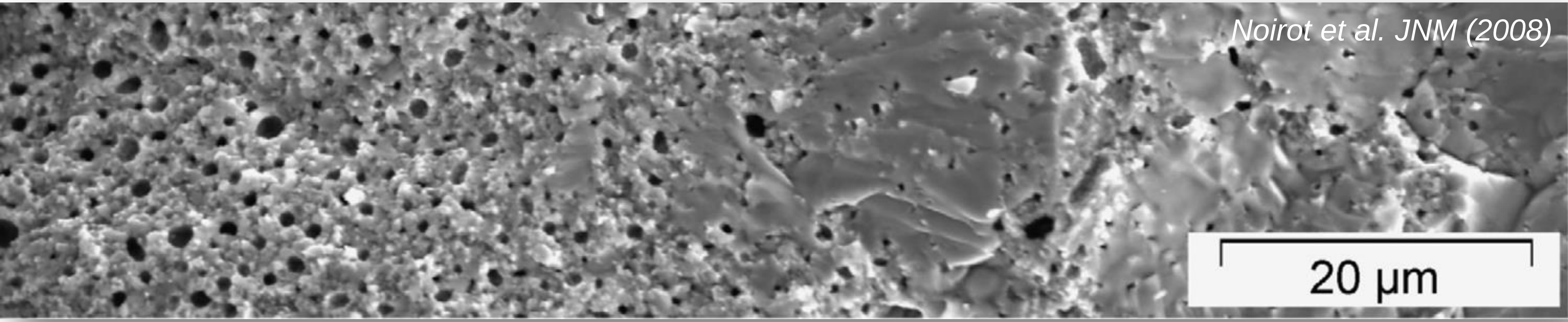
Idaho National Laboratory

Motivation

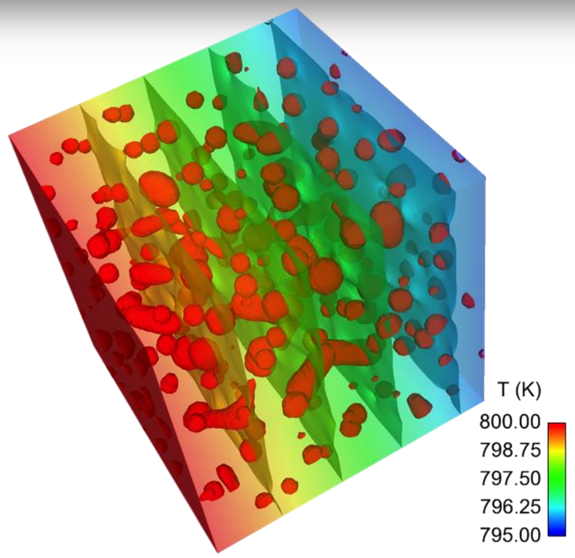
- A coordinated effort to link advanced materials characterization methods and computational modeling approaches critical to understanding and predicting materials behavior in extreme environments
- Difficulty and expense of working with nuclear materials and irradiated materials has inhibited the use of modern characterization techniques
- One example is the reconstruction of 3D digital microstructure which facilitates the application of multi-physics codes such as MARMOT for prediction of important properties such as thermal conductivity, temperature profiles in the microstructure; 3D microstructures are necessary to validate computer models
- FIB methods have been used at INL for 3D reconstruction of microstructure but the area examined is small and the process is time-consuming
- Techniques used for sample preparation must be amenable for use in hot cells

Developing Improved Reactor Materials

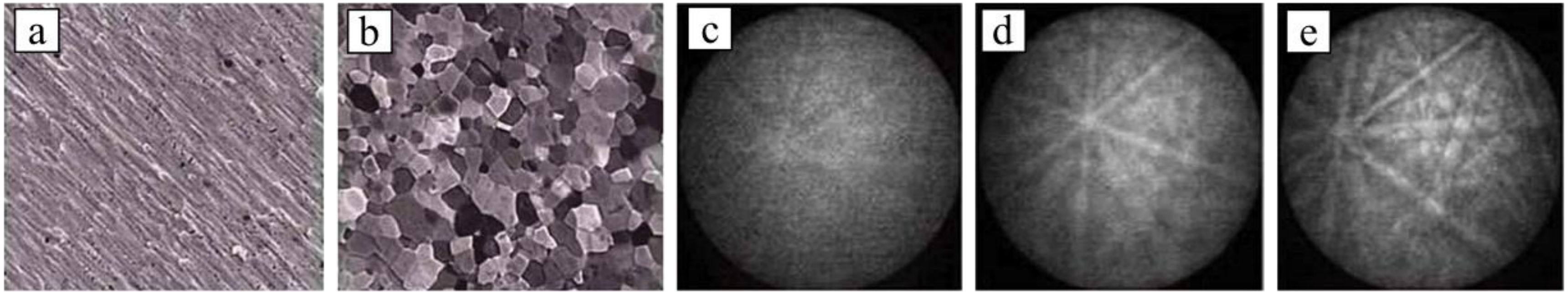
- Safety and efficiency issues in current LWR reactor materials are due to problems with the material behavior



Designing advanced materials for next generation reactors requires a **fundamental understanding** of the **material behavior** of the fuel at the **micron-scale** using **experiments and simulation**.



Broad Ion Beam Etching



Effect of ion milling on titanium with mechanically prepared surface (a) and ion-milled surface (b); The electron backscatter patterns in (c), (d), and (e) represent etch times of 0, 5, and 15 minutes, respectively, for a copper sample [8]

- **Utilizes argon (or xenon) ion to "gently" polish/etch surface of sample**
- **Allows for full cross sections of cladding to be prepared**
- **Hands off technique (i.e. no dose during polishing)**
- **Performed in inert environment, oxidation minimized**
- **Some systems allow application of thin carbon coating while still under vacuum, can "passivate" surface for transfer to EBSD system**

Materials being Investigated

- Unirradiated and radiated HT-9 steel (FFTF, 100dpa): INL
- Unirradiated and radiated 304 stainless steel (ATR, 6dpa): UW-Madison, INL/ATR
- Unirradiated and radiated Zircaloy-4 (BR-3 Reactor, 45 GWd/tHM)

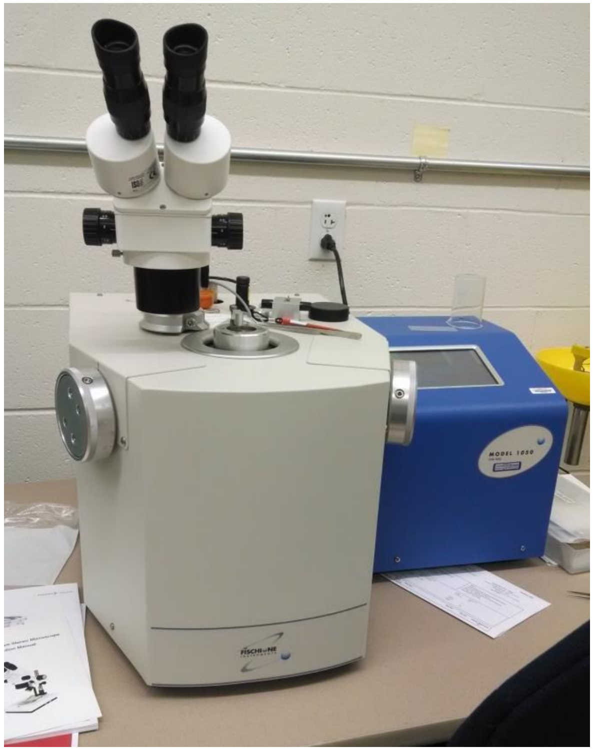
Some unique "Challenges"

- Working with high radiation samples that must be bulk prepared in hot cell introduces some challenge

Parameter	Ideal/Typical	What we can do
Sample Geometry	Free standing, or in very small mount	Must be embedded in mount, hot cell equipment can only produce 1 and 1.25" samples
Sample Height	Shorter the better, maximum 0.75"	Have to be in met mount, hot cells typically produce 0.75" tall mounts
Sample Finish	Colloidal silica vibro-polished, or at "worst" 0.05 micron diamond	Sample finish from hot cell is questionable at best, use diamond paste but finish is significantly worse than out of cell

Ion Etching Equipment

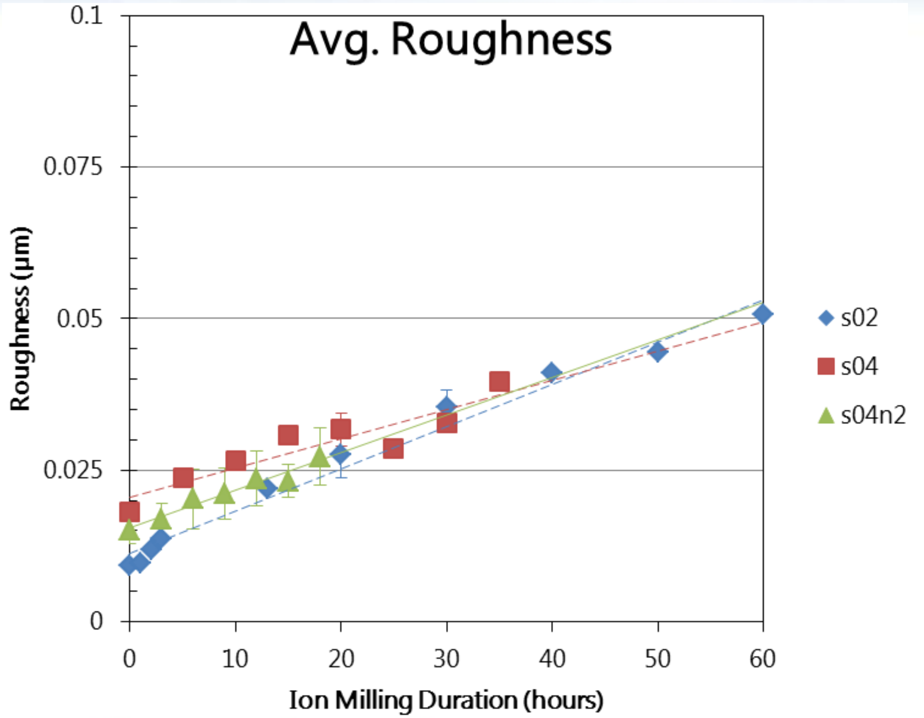
- INL
  - Rad-ready, broad-beam Gatan Model 682 PECS
    - 1-10 keV ions
    - 7-10 mm² etching area (accepts 1.25" met-mounts / SEM stubs)
    - 0-10 mA/cm² current density
- UW Madison
    - Fischione 1050 TEM mill
      - 0.1 – 6 keV
      - <= 10 mA/cm²
      - Have adapted it to polish met mount sized samples
- Collaborating with Gatan on other mills, including development unit

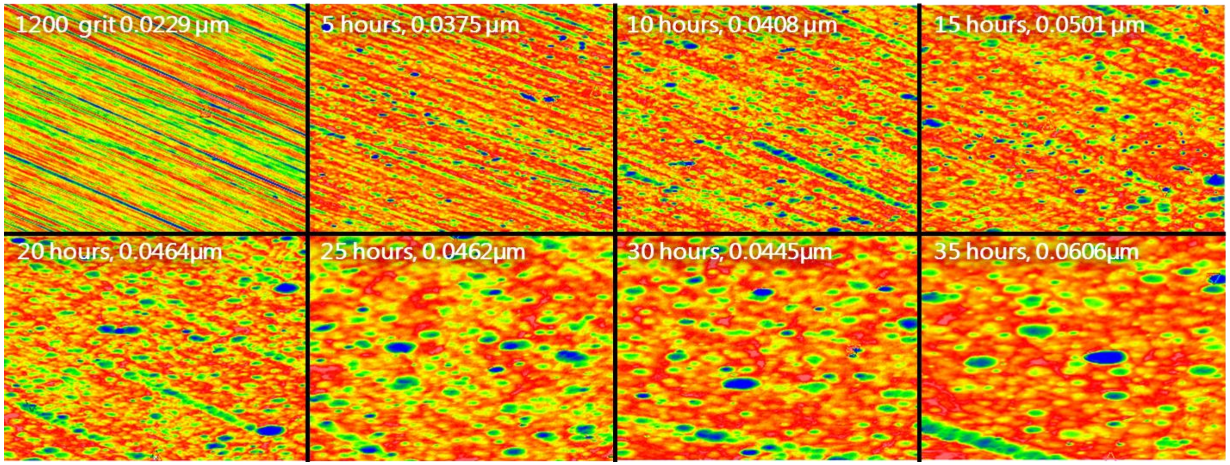


Fischione 1050 TEM Mill at UW-Madison

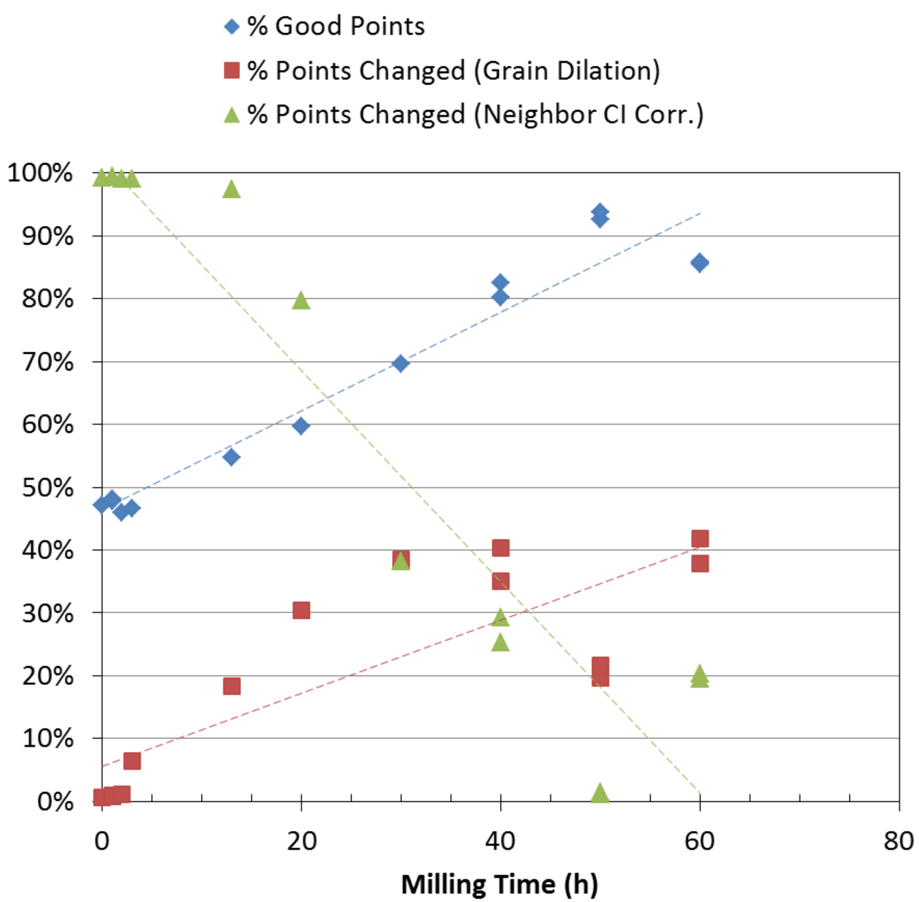
Milling of HT9

- UW-Madison using surface profilometer to measure surface roughness of sample
- Theory was that increased milling would result in decreased roughness, and increased EBSD quality
- Profilometer would provide faster means of measuring results of milling
- Found that roughness actually increases with increased milling time

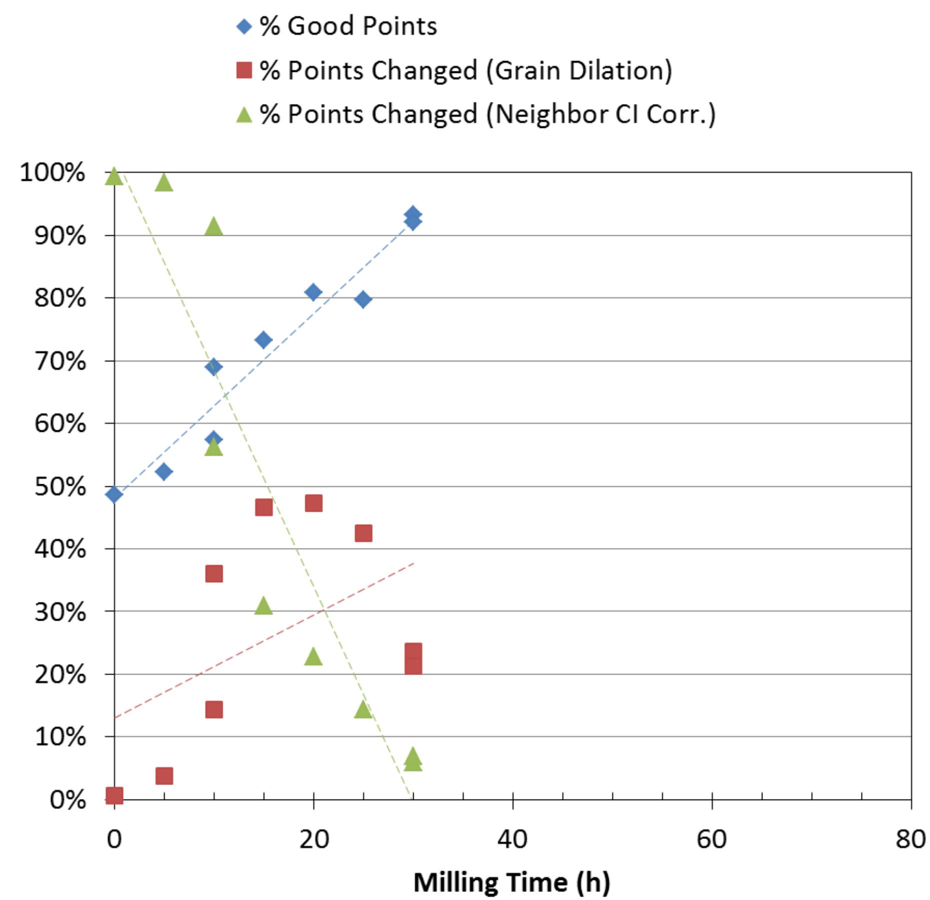




Effect of Sample Height

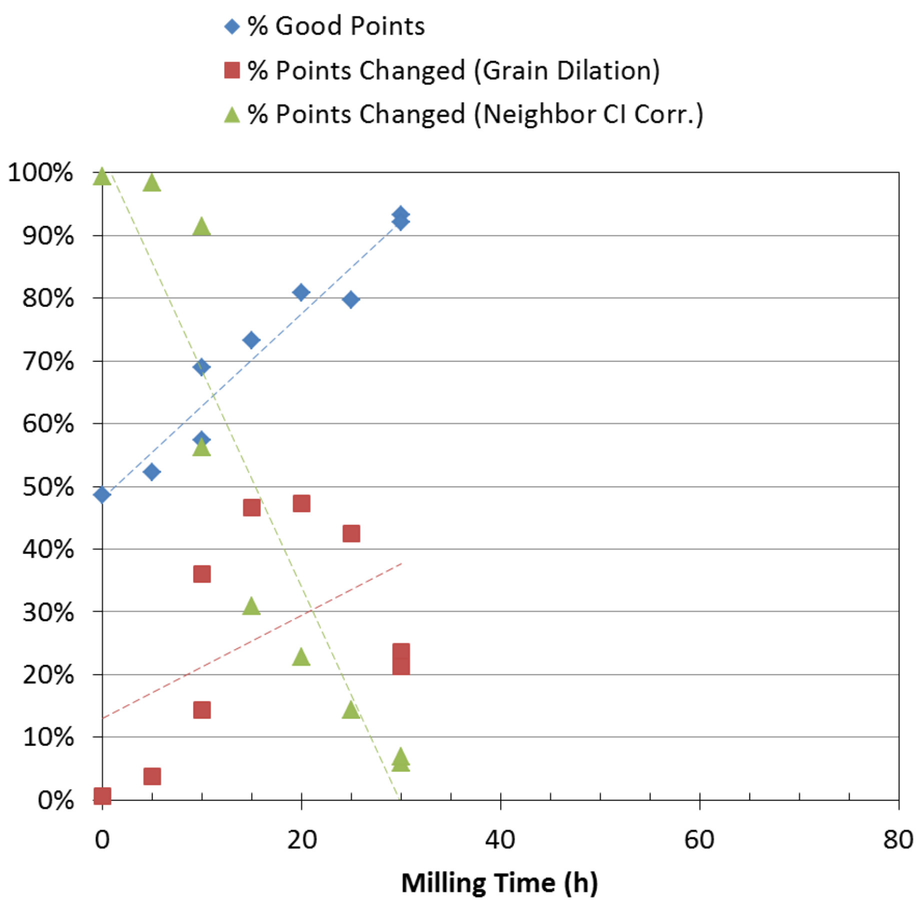


HT9r01s02 (taller)

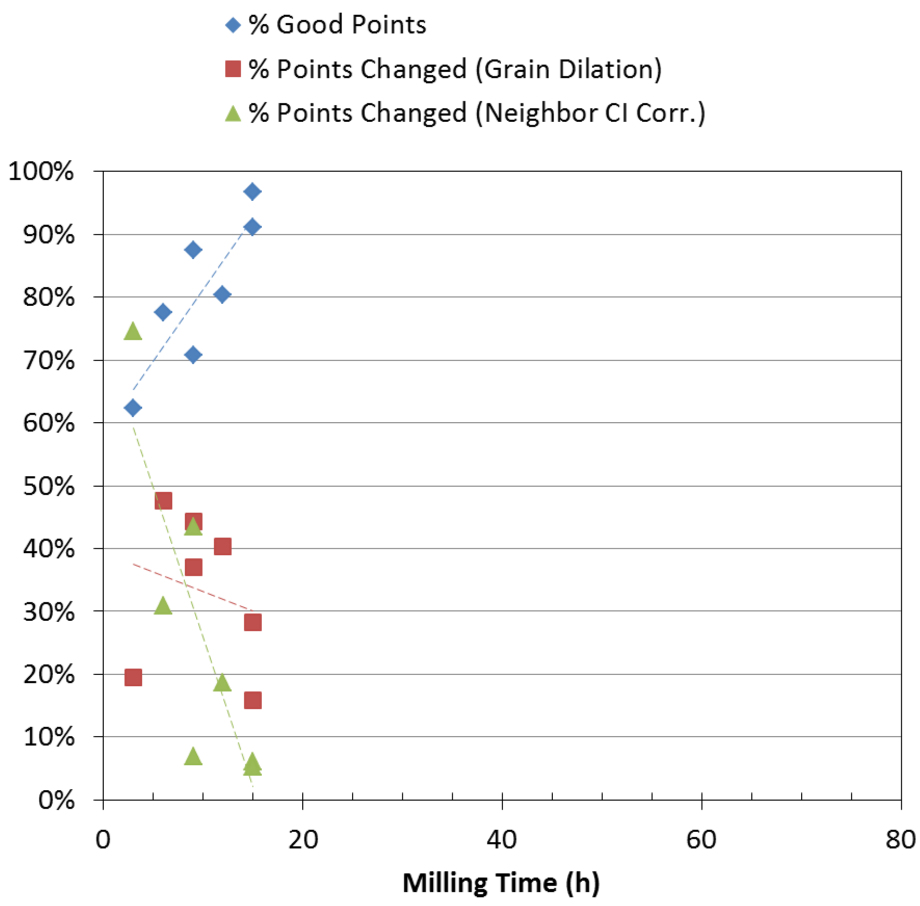


HT9r01s04 (shorter)

Effect of Accelerating Voltage

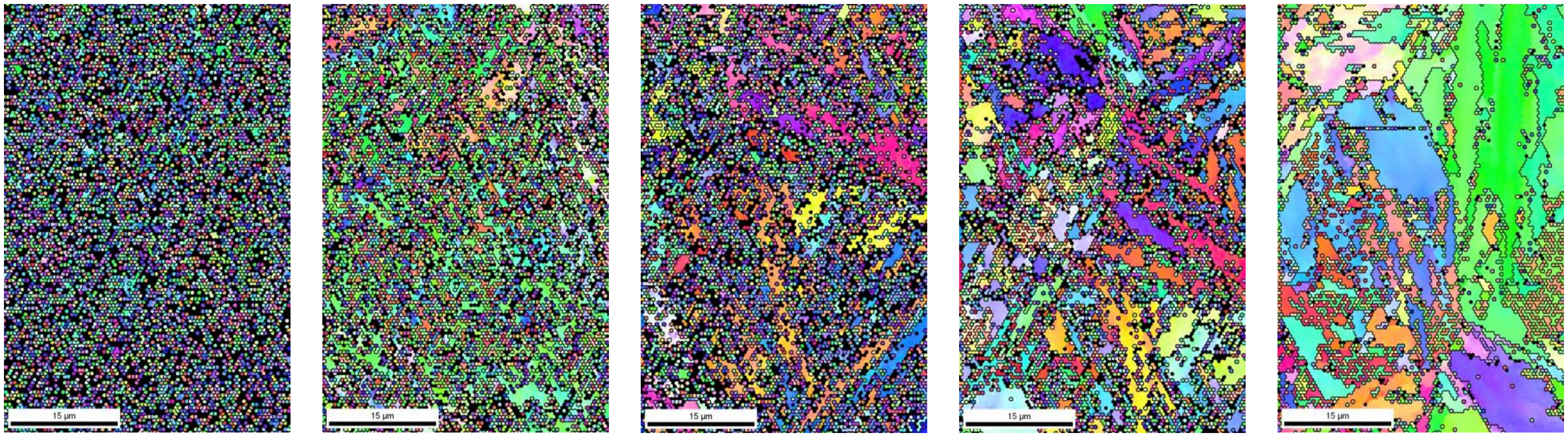


S04
(Trial 3, 3kV only)

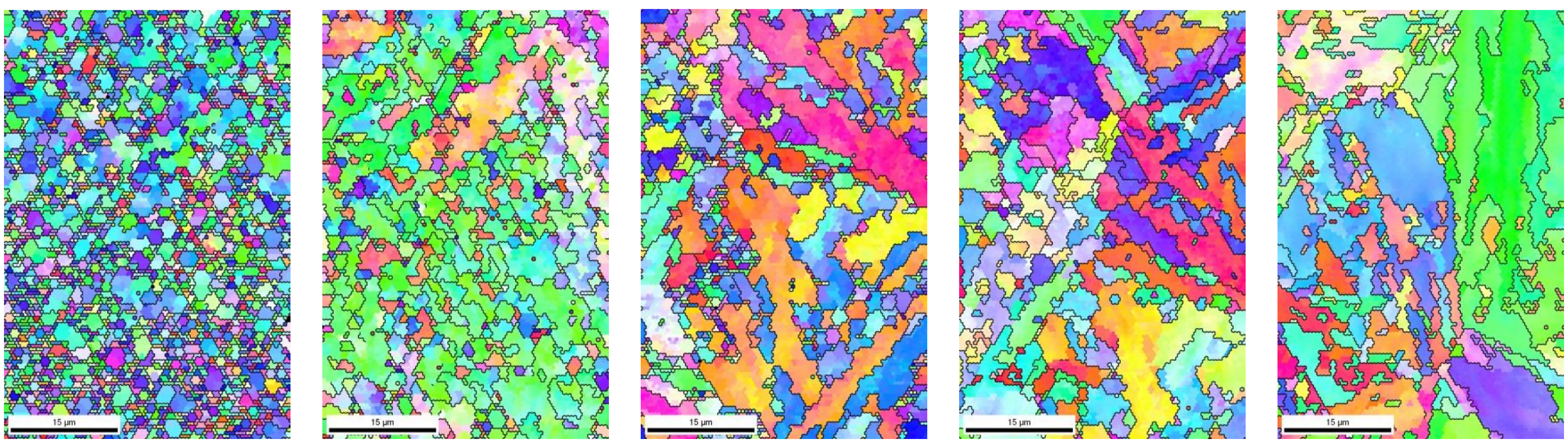


S04n2
(Trial 4, 3h6kV + 1h3kV cycles)

EBSD Improvement with extended time



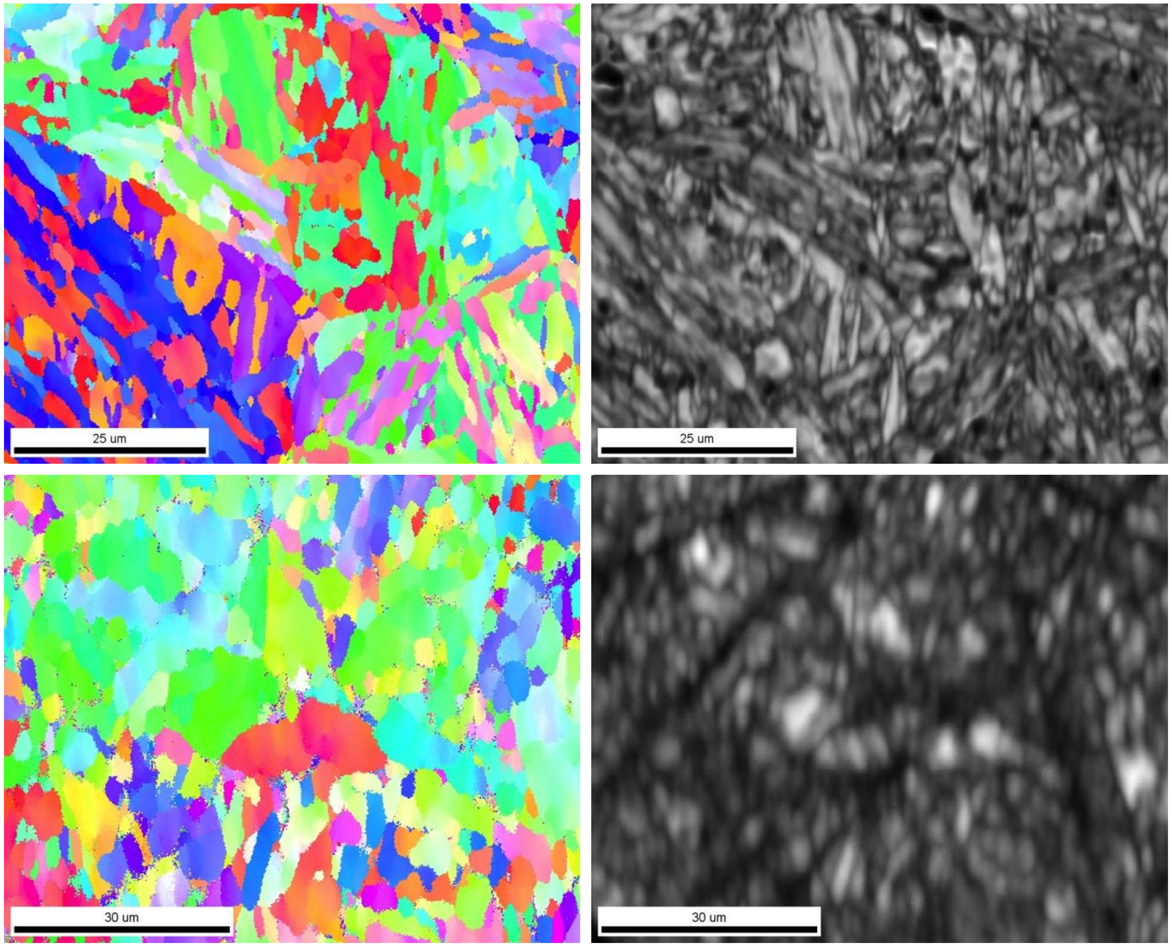
(3h@6kV + 1h@3kV) x 1 | (3h@6kV + 1h@3kV) x 2 | (3h@6kV + 1h@3kV) x 3 | (3h@6kV + 1h@3kV) x 4 | (3h@6kV + 1h@3kV) x 5



As Collected

Processed

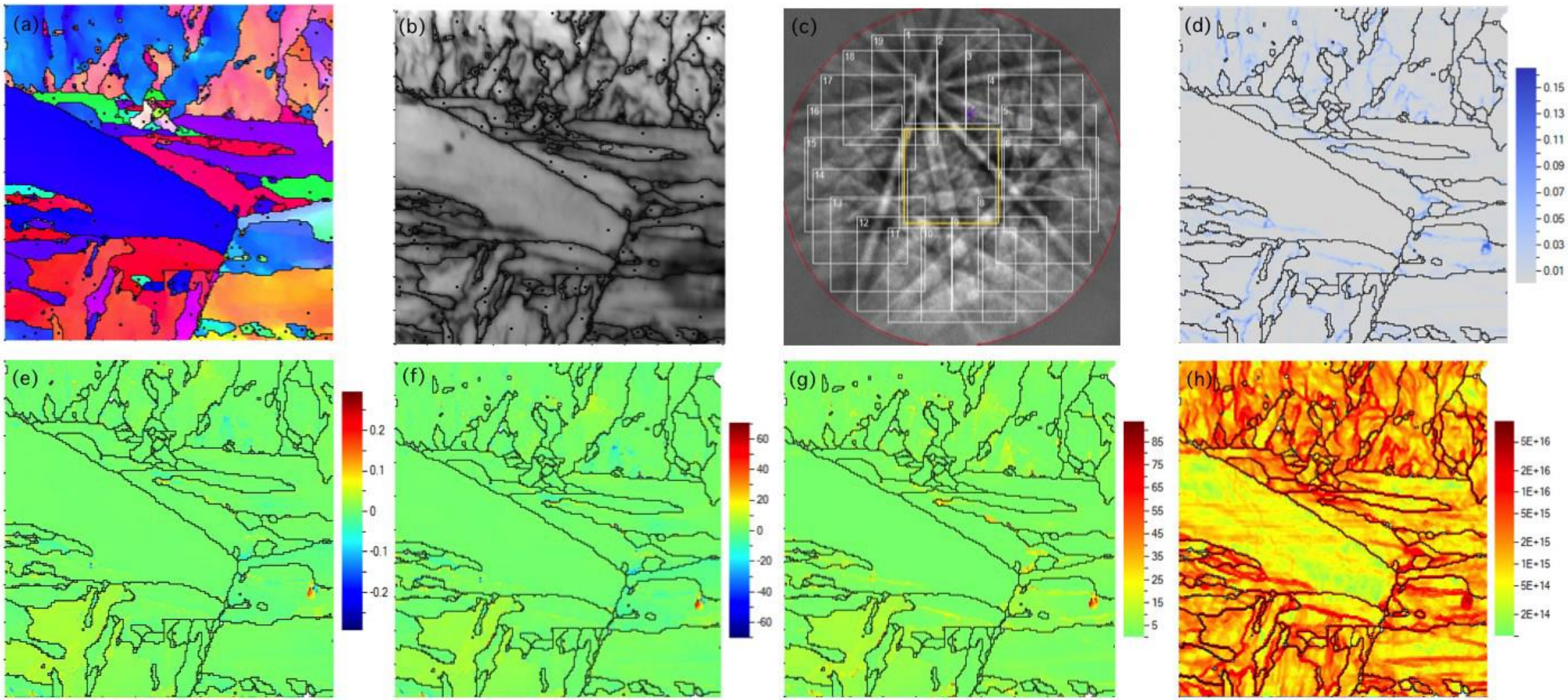
EBSD of HT-9



broad-beam ion etching

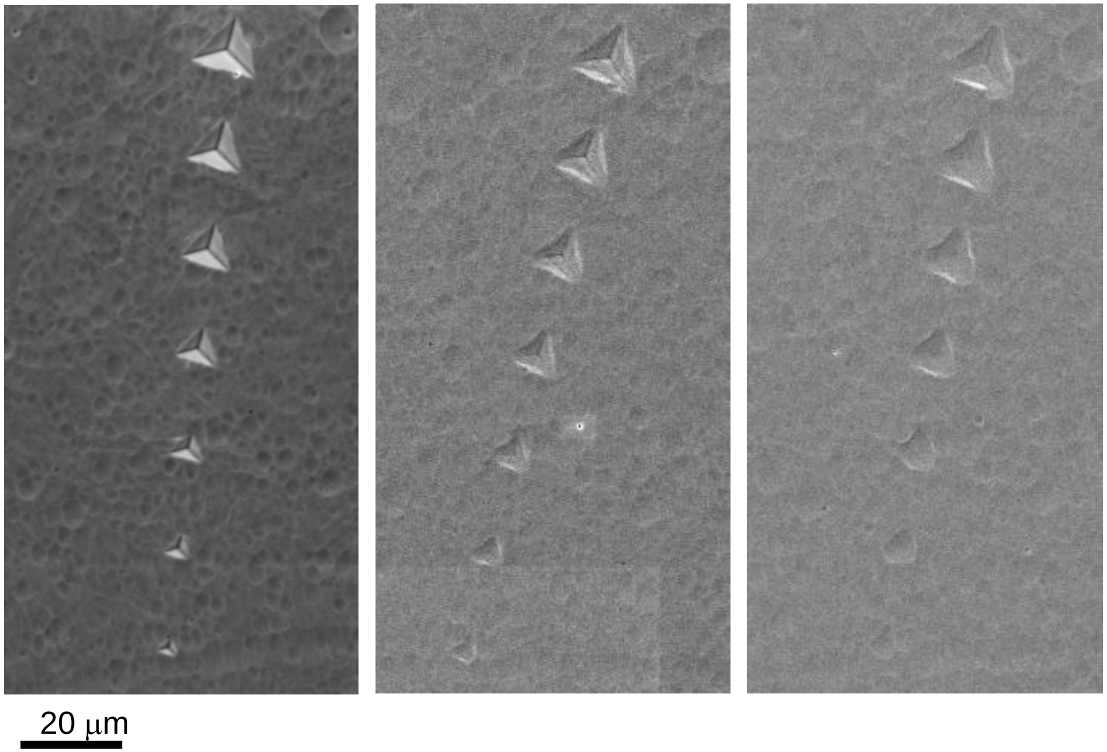
traditional polishing method

HR-EBSD of unirradiated HT-9



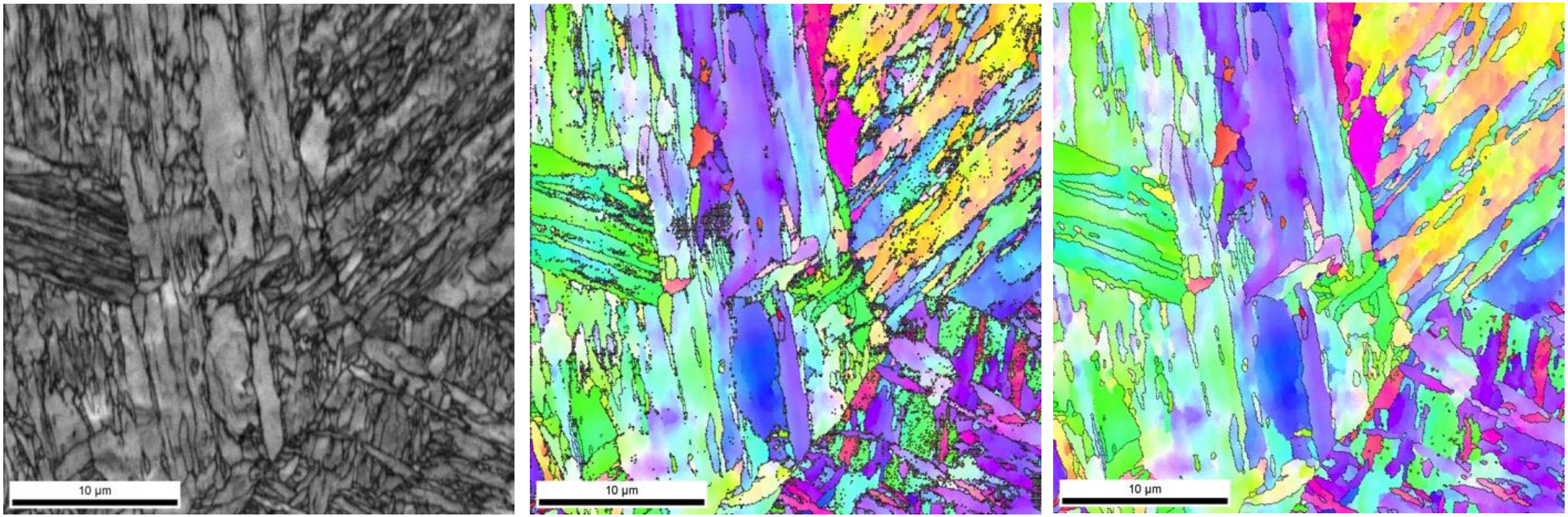
Results from CrossCourt software analysis of HT-9 cladding: (a) inverse pole figure map, (b) image quality map, (c) reference EBSP with 20 regions of interest overlaid onto pattern, (d) high resolution kernel average misorientation map, (e) ε_{22} strain map, (f) σ_{22} stress map, (g) Von Mises stress plot, (h) geometric necessary dislocation map

Serial sectioning of unirradiated HT-9



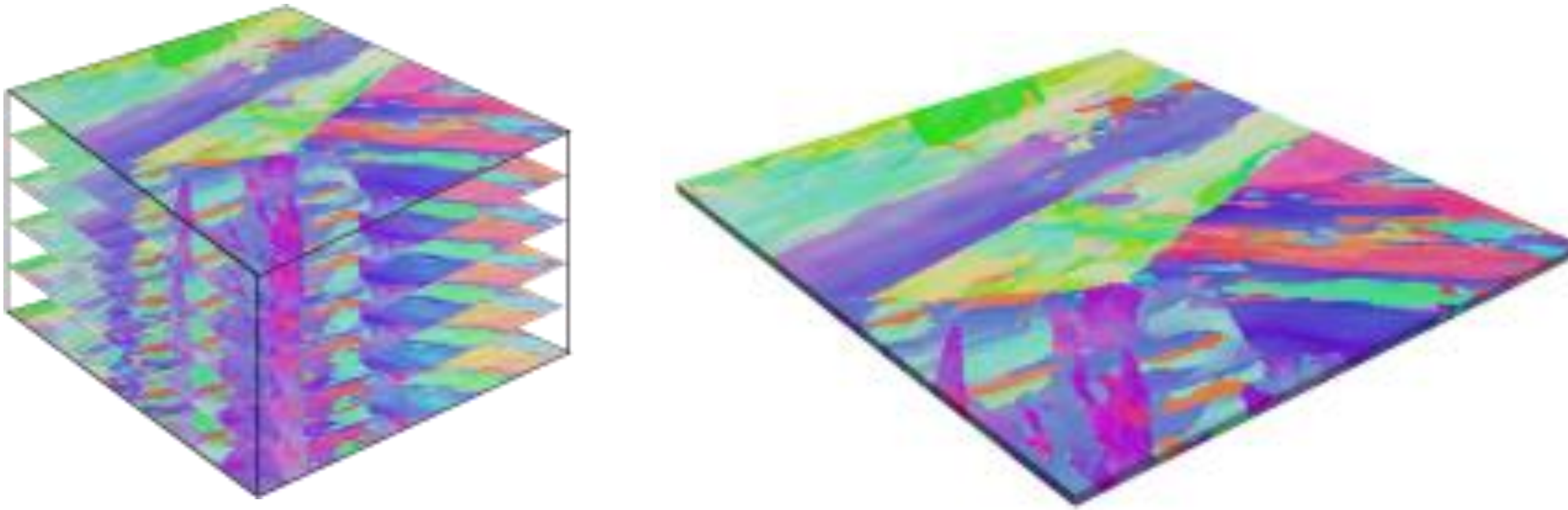
Milling rate analysis of HT-9 sample prepared with the PECS. The image on the left shows the 7 nanoindentation marks prior to etching. The middle image was recorded after 1.5 hours of milling at 4.5 kV. The right image represents 3 hours of milling. The milling rate was estimated to be ~100 nm per hour at 4.5 kV.

Serial sectioning of unirradiated HT-9



EBSD results for Slice 1; (a) Image Quality map; (b) Inverse Pole Figure before cleanup, (c) Inverse pole figure after cleanup.

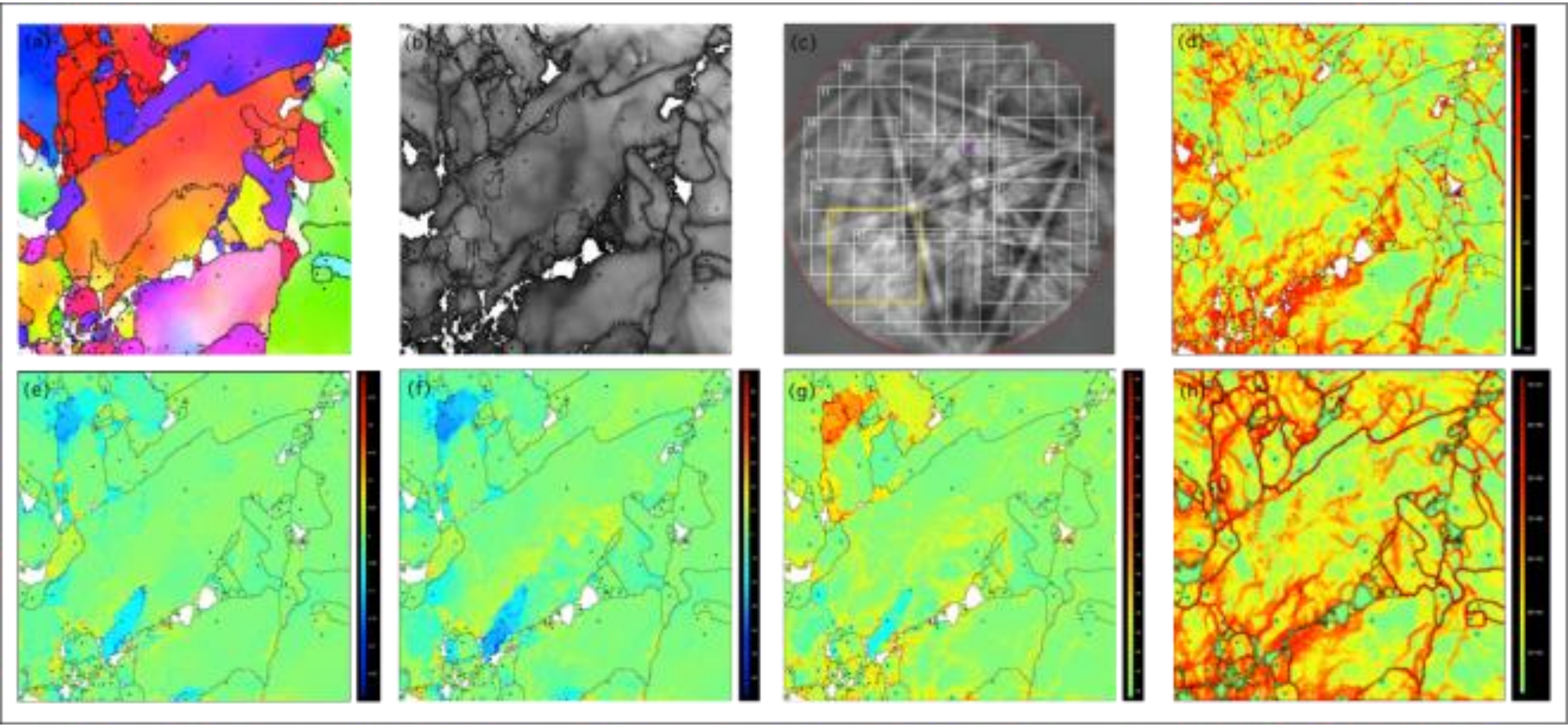
Serial sectioning of unirradiated HT-9



a) 3D EBSD reconstruction of HT-9 showing microstructure of the material and b) Serial EBSD sections aligned in a stack prior to the reconstruction.

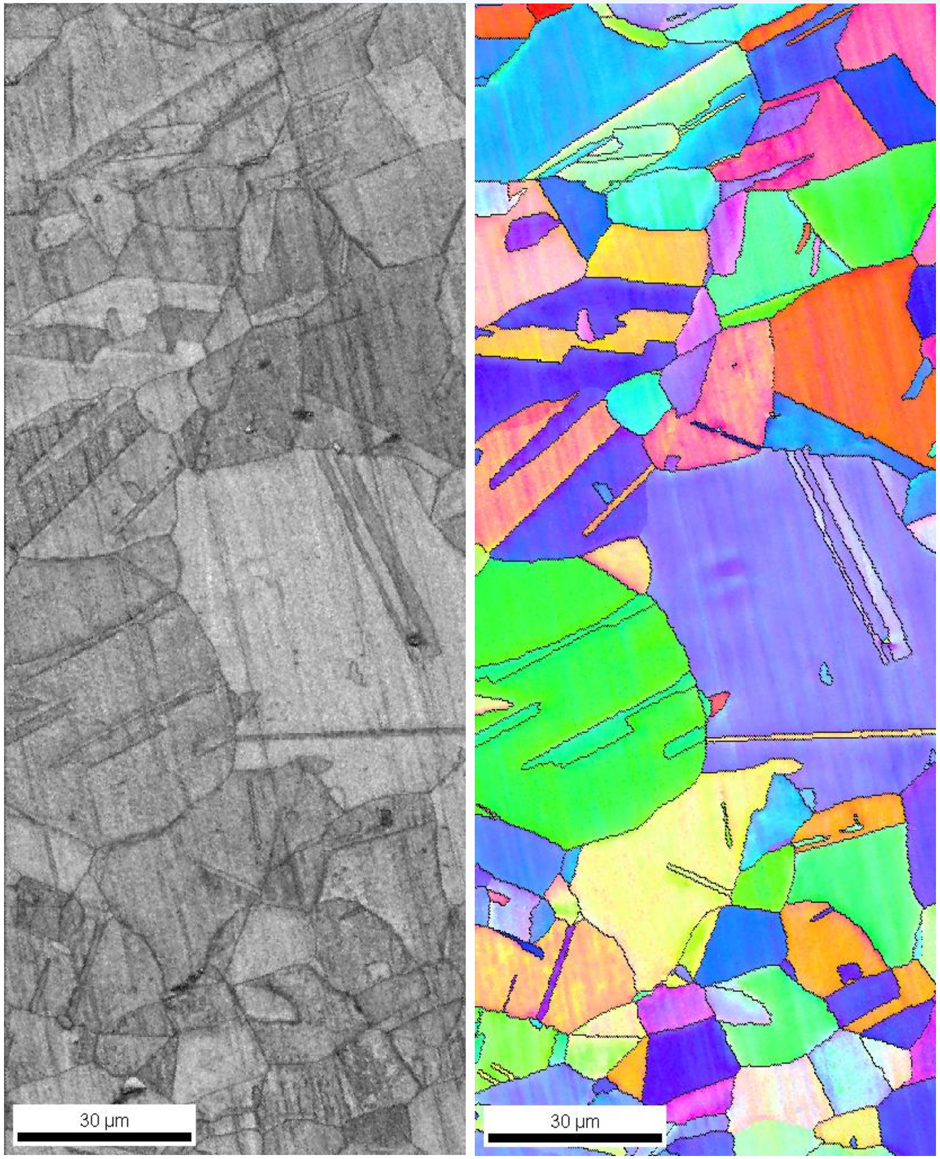
0.375 microns total depth

HR-EBSD of irradiated HT-9



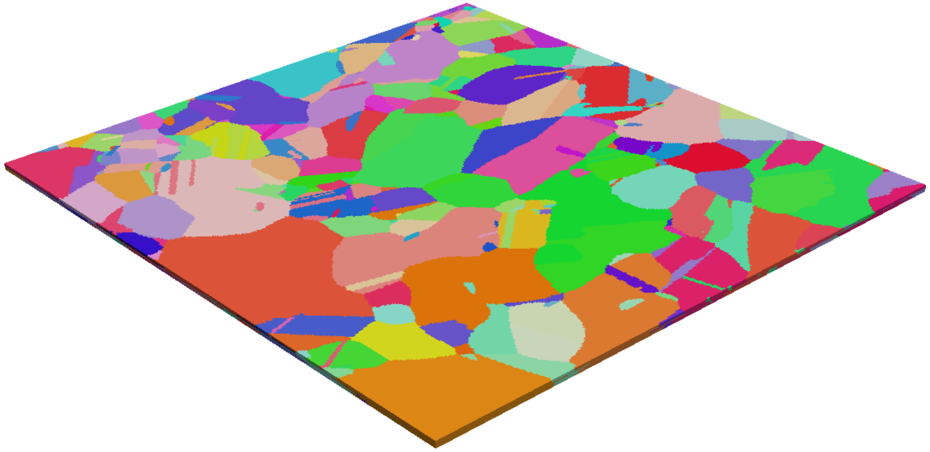
Results from HR-EBSD analysis of irradiated HT9 cladding: (a) IPF map, (b) IQ map, (c) reference electron backscatter pattern, (d) HR-KAM map, (e) strain map, (f) stress map, (g) Von Mises stress plot, (h) geometric necessary dislocation map.

EBSD of unirradiated 304H



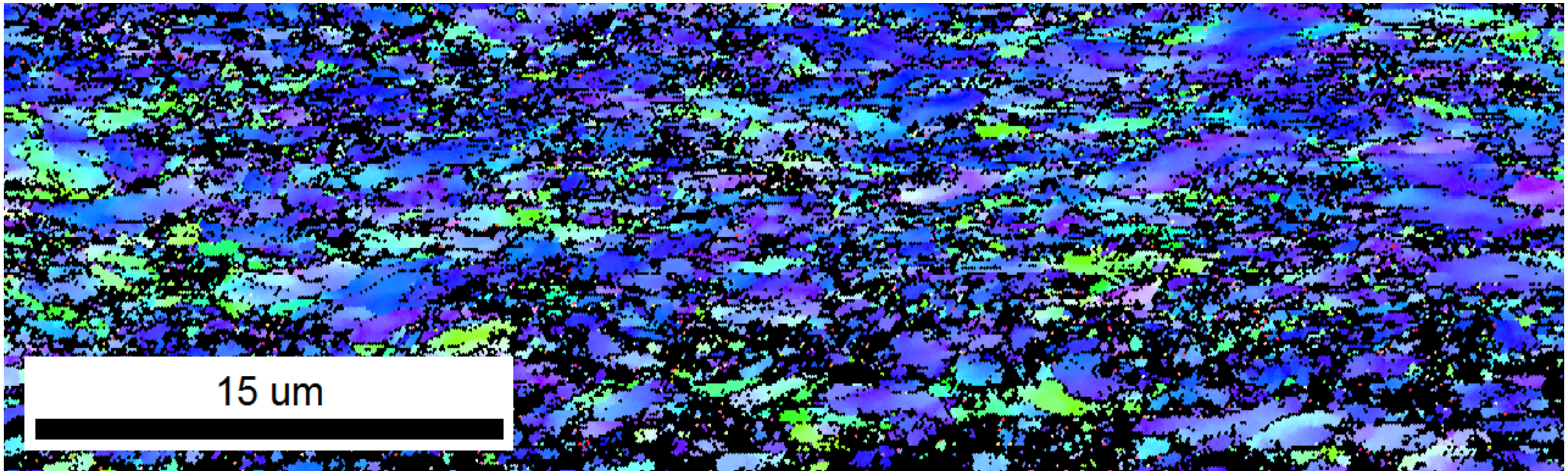
Results from EBSD analysis of 304H steel: (a) IQ map and (b) IPF map.

EBSD of irradiated 304H



2 microns total depth

Ion Milling Zr-4 Cladding



- All points show >0.1 CI with no cleaning
- Able to achieve EBSD pattern with higher quality than achieved with best mechanical polishing
- Sample “deteriorates” quickly due to oxidation
- Experimenting with adding a thin carbon coating in PECS to passivate surface

Summary

- Broad beam etching (milling) being investigated for surface polishing/etching of ‘hot’ samples in a hot cell environment to provide high quality EBSD images
- Optimization of milling parameters to perform serial sectioning so that EBSD patterns can be obtained at various known depths that can be used for reconstruction of 3D grain structure that can be used for MARMOT simulations
- Work has been successfully completed for unirradiated and irradiated HT-9 ferritic-martensitic steel and super 304 stainless steel
- Work on Zr-alloy cladding material is in progress, but this material presents challenges on account of its propensity to oxidize which deteriorates EBSD patterns
- Working closely with company Gatan in testing and developing new ion etching systems to expand capabilities towards reducing milling time and improving EBSD pattern quality

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