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Investigation of High Burnup Ceramic Fuel Microstructure at Idaho National Laboratory

Fabiola Cappia

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Investigation of High Burnup Ceramic Fuel Microstructure at Idaho National Laboratory

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Outline

- **Introduction**
  - The High Burnup Structure (HBS)
  - Microstructural modifications due to irradiation: implications for fuel performance

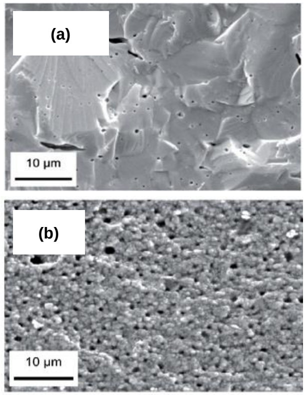
- **Results highlights**
  - High burnup commercial fuel advanced microscopy
  - Separate effect testing: the role of initial microstructure in HBS formation

- **Summary**

Irradiation-induced changes

- Thermo-physical properties
- Dimensional changes
  - Fuel Swelling
  - Axial Growth
  - Cladding creep
- Fission gas production and release (pin pressure)
- Fuel restructuring (central hole formation), cracking, densification
- Constituent redistribution
- Fuel cladding chemical/mechanical interaction
- Cladding/materials corrosion
- **High Burnup Structure (HBS) formation**

Original structure



High Burnup Structure

Local enrichment in ^{239}Pu at pellet periphery takes place

↓

1. Higher local fission density, high radiation damage

+

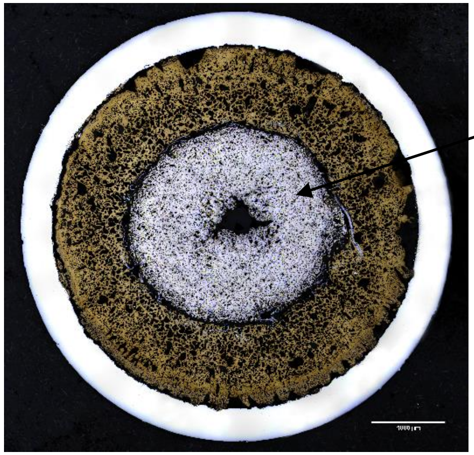
2. Limited thermal recovery

=

A modified structure is formed at the pellet periphery

SEM micrographs showing fuel fracture surfaces: (a) unirradiated fuel; (b) fuel with local burn-up of ~75 GWd/tHM

U-10Zr AFC campaign

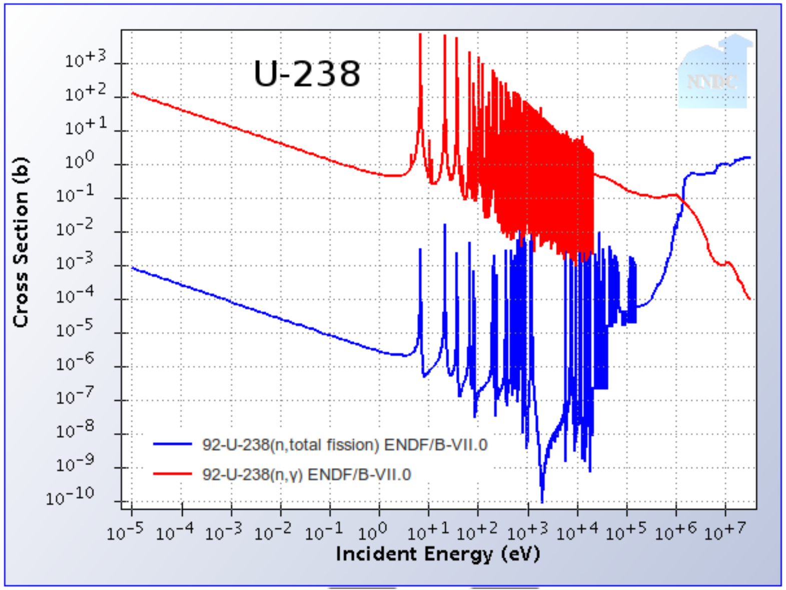


Zr-rich phase

Radial redistribution of components

U-10Zr, 3.3% FIMA

Microstructural modifications: High Burnup Structure (HBS)



$$^{238}_{92}\mathrm{U} + ^{1}_{0}n \longrightarrow ^{239}_{92}\mathrm{U}$$

$$^{239}_{92}\mathrm{U} \xrightarrow[\text{mins}]{23} ^{239}_{93}\mathrm{Np} + \beta^-$$

$$^{239}_{93}\mathrm{Np} \xrightarrow[\text{days}]{2.36} ^{239}_{94}\mathrm{Pu} + \beta^-$$

$$^{239}_{94}\mathrm{Pu} \xrightarrow[\text{years}]{24100} ^{235}_{92}\mathrm{U} + ^{4}_{2}\mathrm{He}$$

Local enrichment in ^{239}Pu at pellet periphery takes place

↓

1. Higher local fission density, high radiation damage

+

2. Limited thermal recovery

=

A modified structure is formed at the pellet periphery

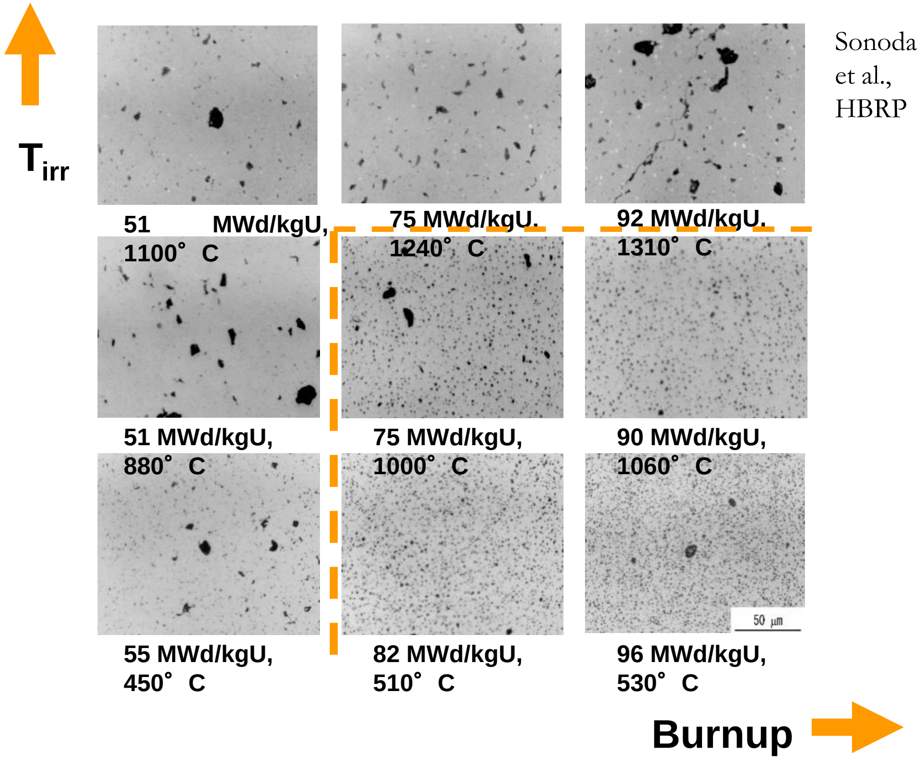
Phenomenon primarily occurring in UO_2 LWR fuels

HBS summary

- **Characteristics**
  - Grain subdivision/recrystallization
  - Formation of micron-sized inter-granular closed pores
  - Depletion of fission gases from the fuel matrix

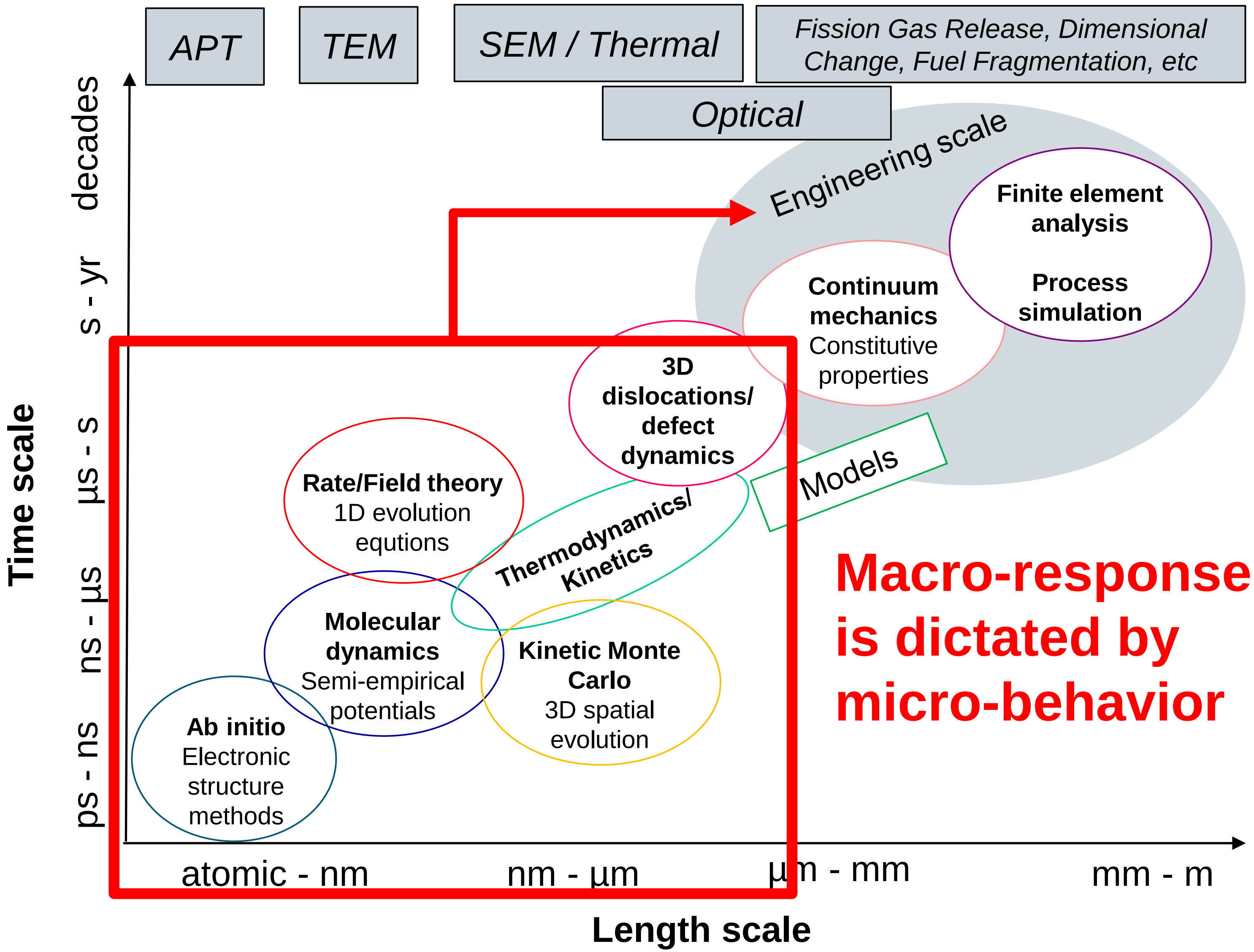
- **Conditions for formation**
  - $T_{irradiation} \leq 1000^\circ$ C (approximately)
  - Local burnup ≥ 50-60 GWd/tHM

- **Large impact on fuel properties (hence, performance)**
  - Mechanical → increased porosity reduces load-bearing area
  - Porosity increase → swelling (stress on cladding) and decreases thermal conductivity



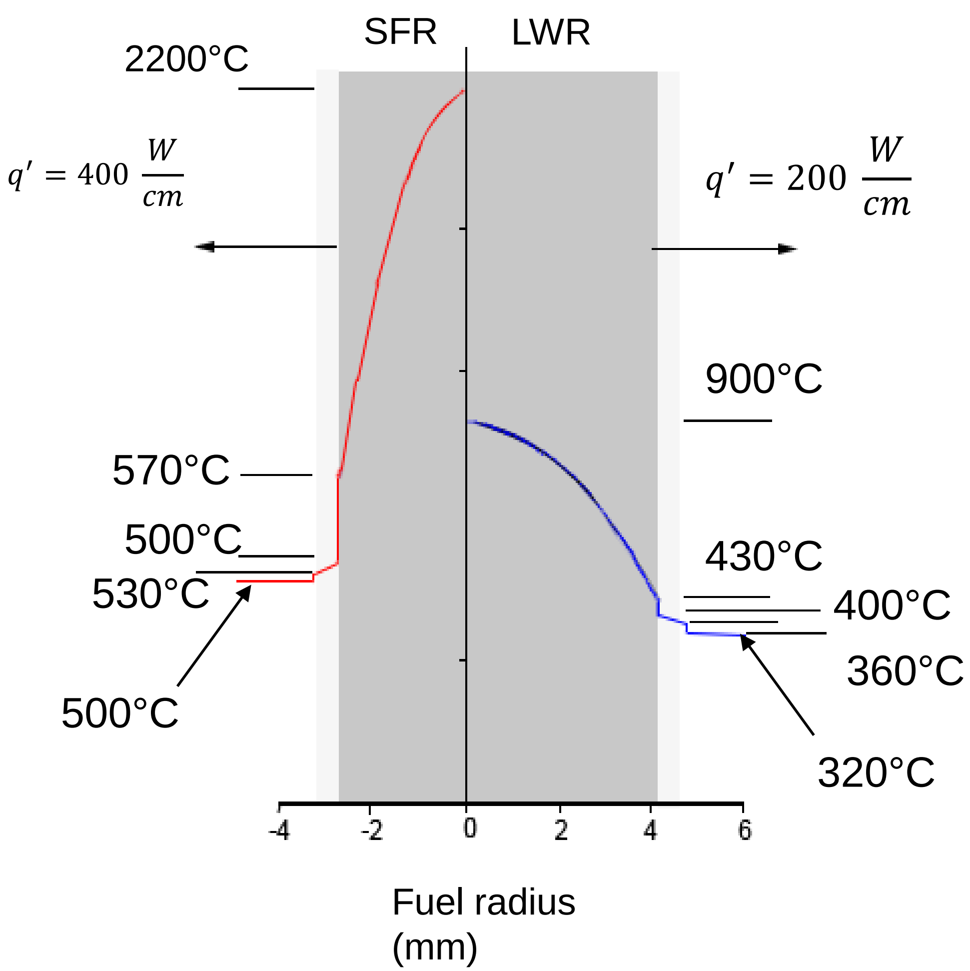
Sonoda et al., HBRP

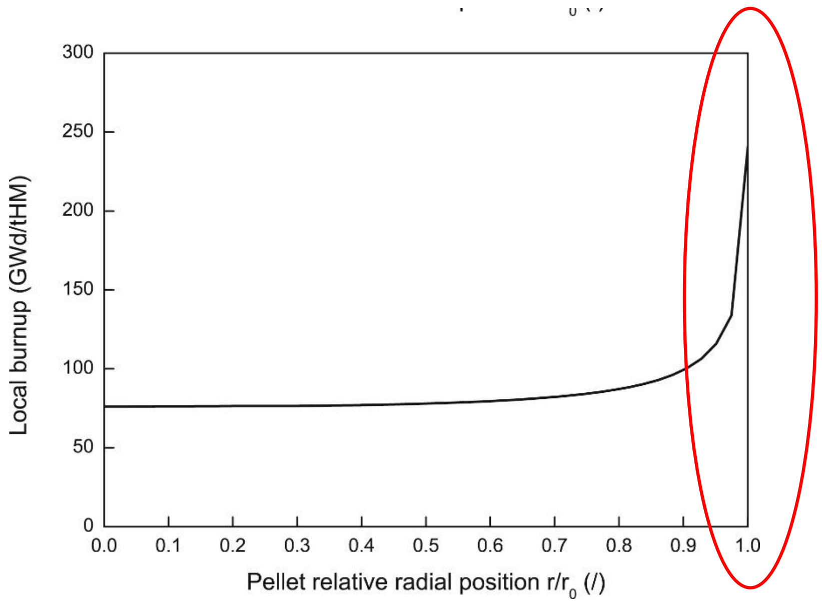
Motivation for multi-scale investigation



Macro-response is dictated by micro-behavior

Challenges in microstructural characterization of nuclear fuels

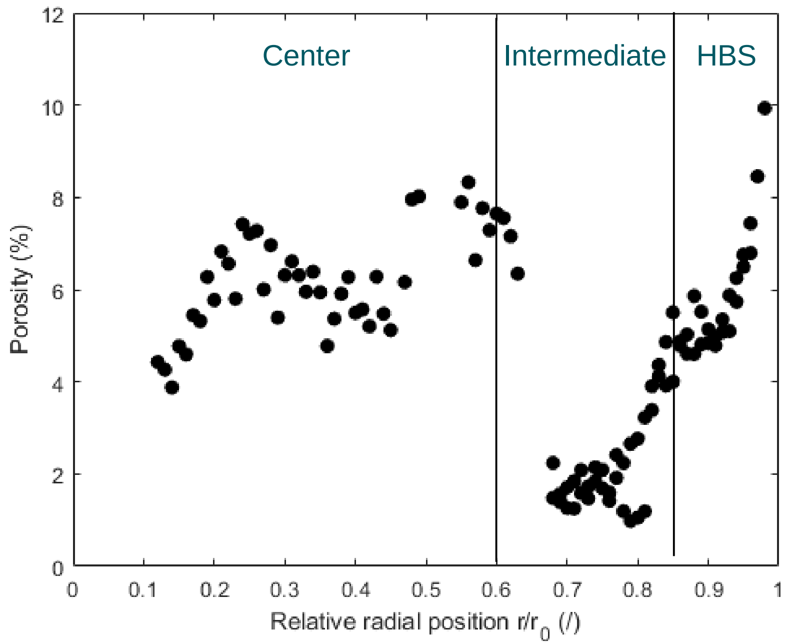


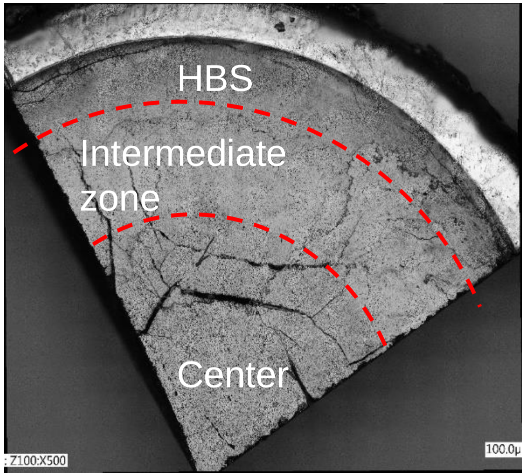


Thermal and burnup gradients require analyses with the same spatial resolution, but keeping statistical significance

Characterization of microstructure along the radius: porosity

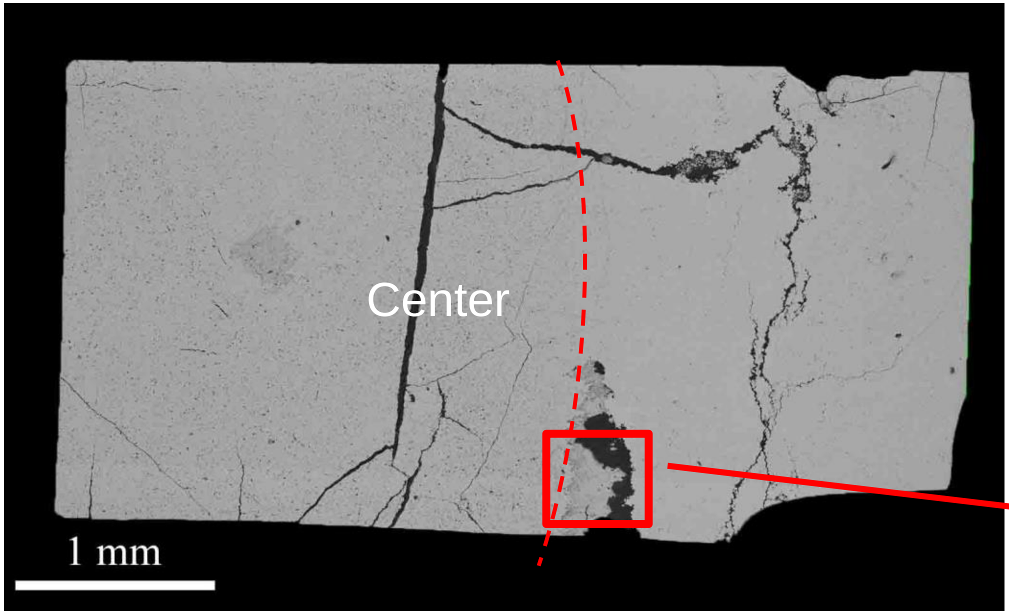
- Advanced microscopy using SEM/FIB/EBSD is the right tool to tackle the radial heterogeneities
- Experimental campaign on a high burnup (rod average 72.9 GWd/tHM) for characterization before TREAT LOCA safety testing

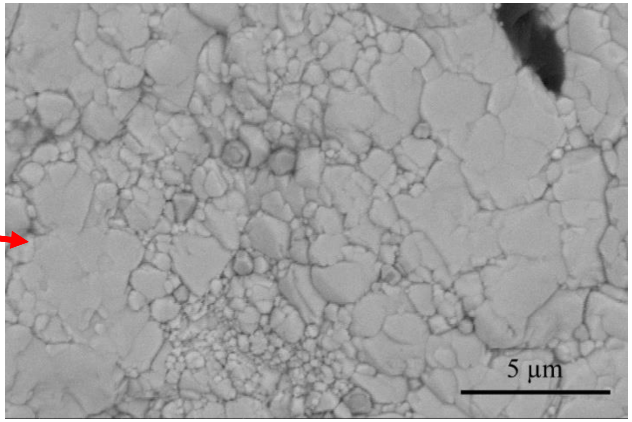




- Porosity quantification highlights what already visible qualitatively: a formation of 3 macro-zones
- No details about the irradiation history are available, but from the porosity we can conclude that the linear heat generation rate (LHGR) was probably high

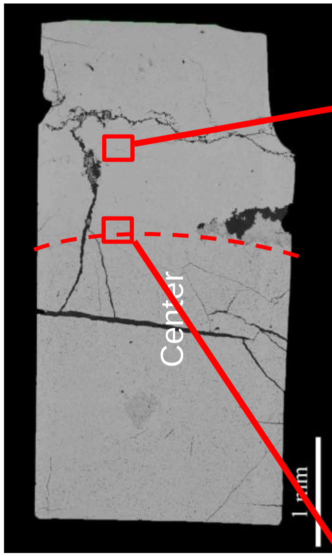
Characterization of microstructure along the radius: SEM and EBSD (1/2)

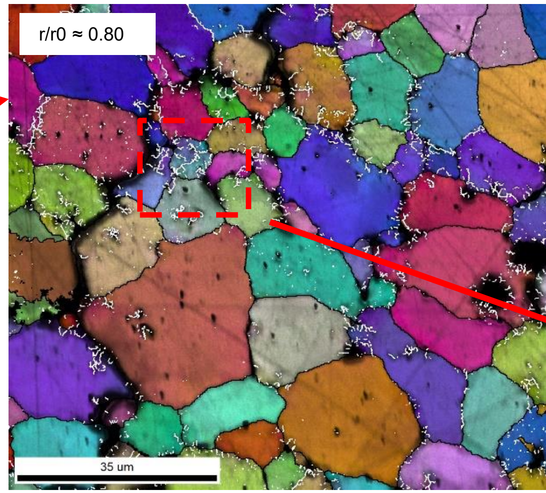




- Restructuring at the pellet periphery is a very well-known phenomenon
- **But also intermediate regions start to develop grain restructuring**

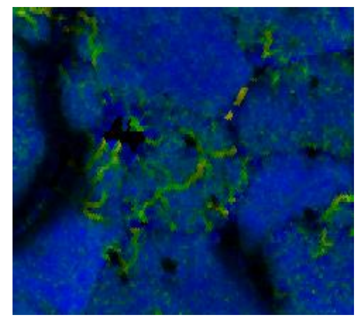
Characterization of microstructure along the radius: SEM and EBSD (2/2)





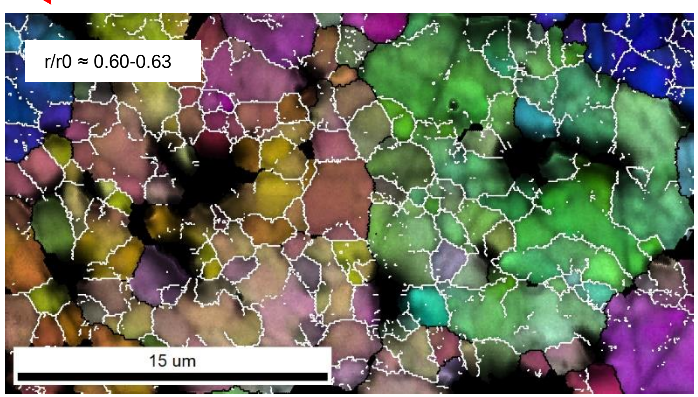
IQ + IPF

- HAGB dominated
- Most LAGB are noise from porosity and around cracking
- Some degree of subdivision might occur starting on grain boundaries (very limited)

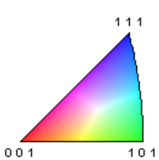


IQ + KAM map < 15º

(white line)	LAGB (2º<x<15º)
(black line)	HAGB (>15º)



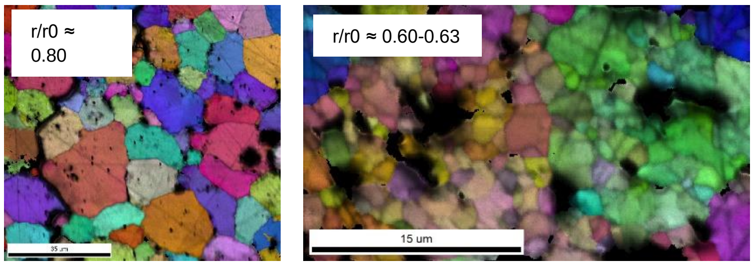
IQ + IPF

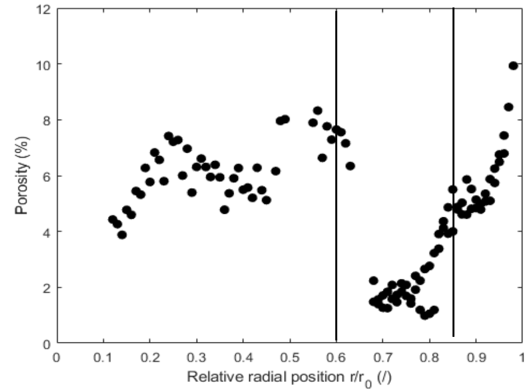


- LAGB dominated
- HAGB are associated with original grain boundaries

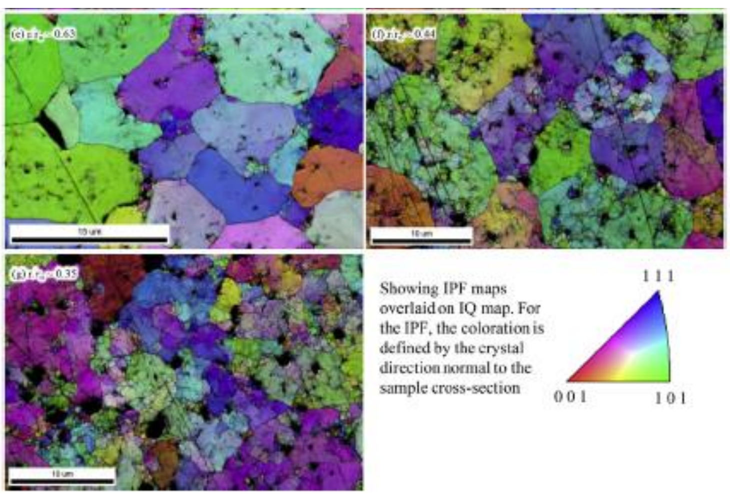
Comparison with another high burnup sample

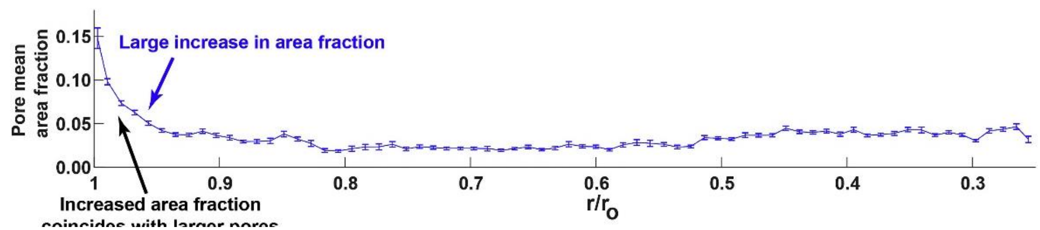
72.9 GWd/tHM





72 GWd/tHM

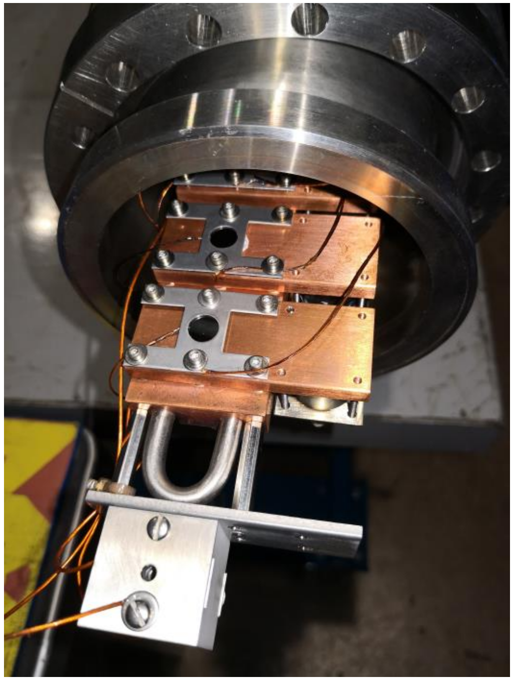




Gerczak et al., JNM 509 (2018), p. 245-259

- Same burnup, but different histories lead to totally different grain structure
- Too many variables are involved to provide a clear understanding of the observations – **separate effect testing needed**

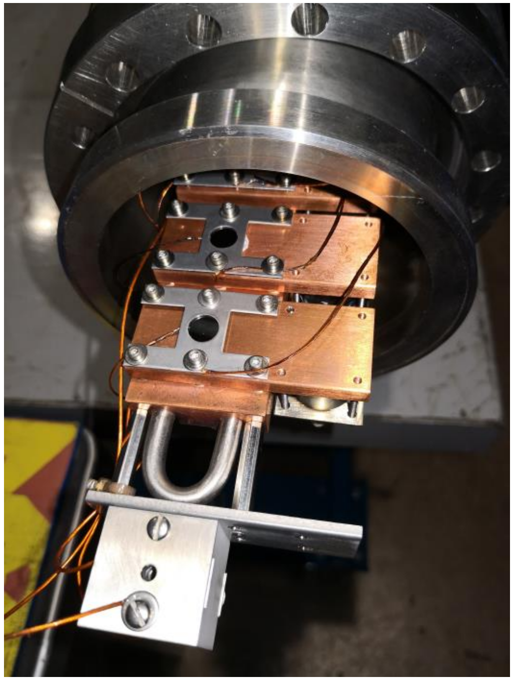
High energy Xe ion-irradiation to understand HBS formation



	Sample A	Sample B
Initial grain size (μm)	10	70
Irradiation temperature (°C)	350	500
Ion fluence (ion/cm^2)	7.22×10^{17}	7.94×10^{17}
dpa (SRIM calculations)	1357	1492.7
% FIMA	5.35	5.88

Samples fabricated at RPI using SPS
Samples irradiated at ANL ATLAS beamline with 84 MeV Xe

High energy Xe ion-irradiation to understand HBS formation (1/4)



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Samples fabricated at RPI using SPS
Samples irradiated at ANL ATLAS beamline with 84 MeV Xe

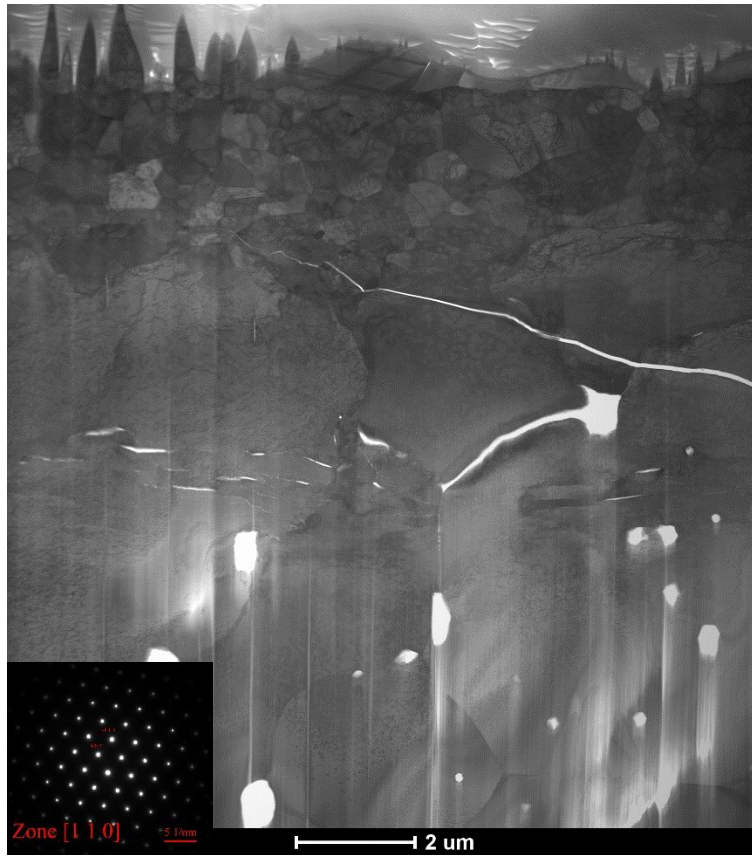
High energy Xe ion-irradiation to mimic HBS formation (2/4)

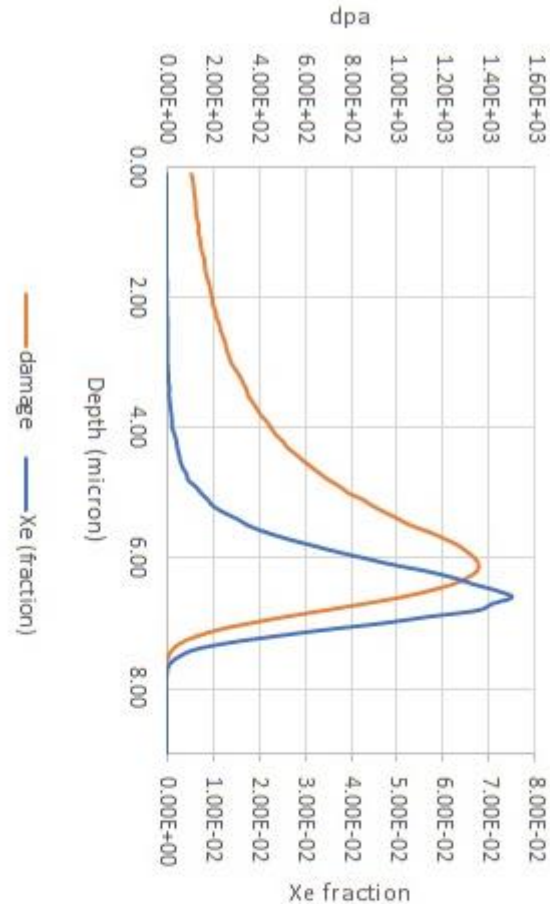
Hillocks

Restructured area with grain subdivision

Intermediate zone with damage gradient

Bubble formation

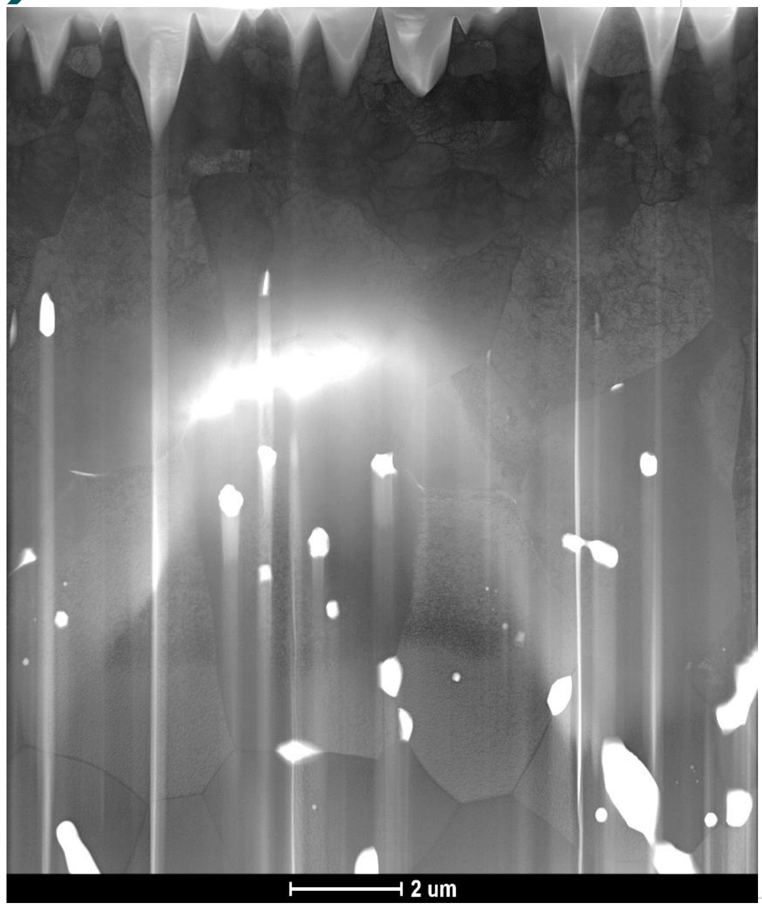




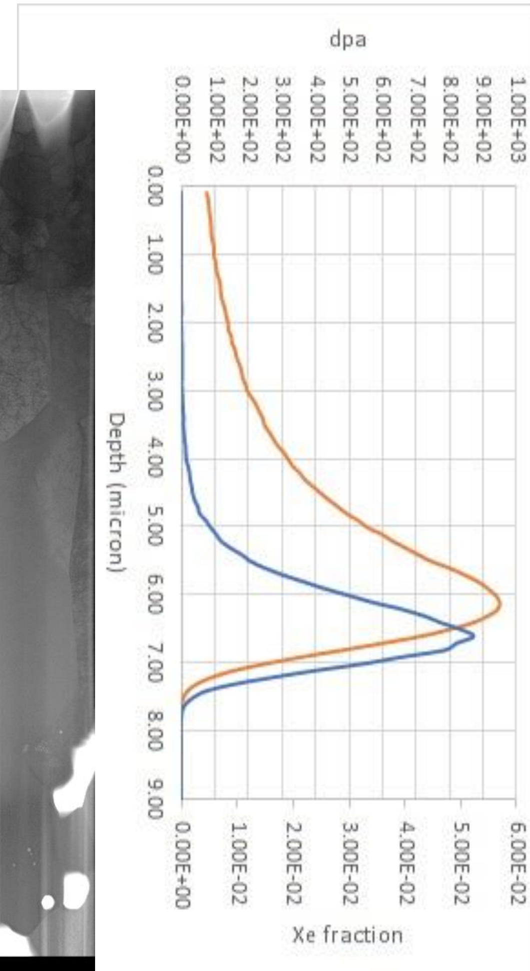
Sampled at peak beam fluence (7.22×10^{17} ions/cm^2)

High energy Xe ion-irradiation to mimic HBS formation (3/4)

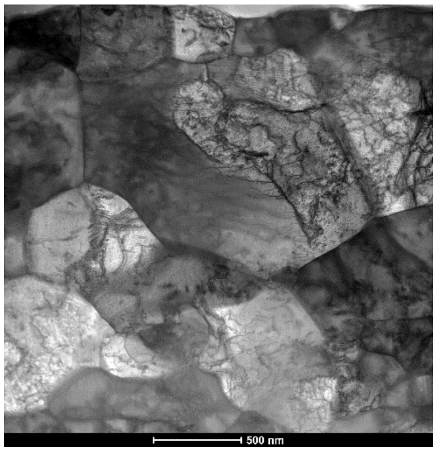
- Hillocks
- Grain subdivision
- Intermediate zone with damage gradient
- Bubble formation



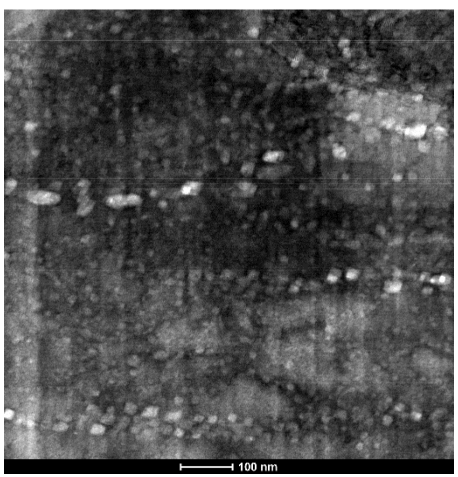
Sampled at ≈70% peak beam fluence (5.05 x 10^{17} ions/cm^2)

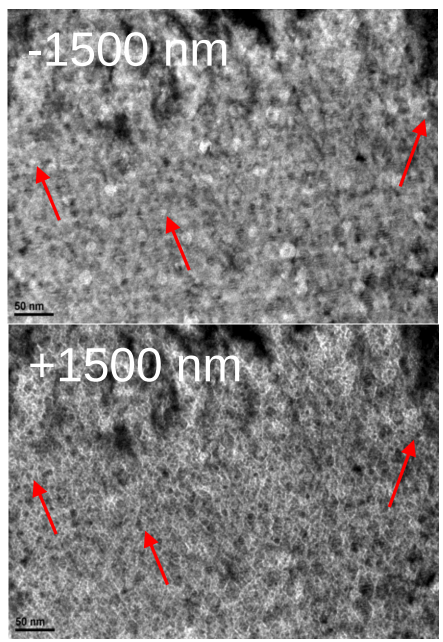


High energy Xe ion-irradiation to mimic HBS formation (4/4)



- Grain polygonization in UO_2 by swift heavy ion irradiation already reported by Wiss et al. in *NIMB* (2000)
- At the Xe peak deposition, 2 classes of bubble size observed. Results consistent with previous work by Miao et al. in *Scripta Materialia* (2018)





- **Grain subdivision was not observed at the peak dose area, rather it occurs independently from the formation of bubbles**
- Restructuring occurs at the sample surface, where electronic energy losses dominate
- Real effect or influence of sample oxidation? More characterization planned

Summary

- Modern characterization techniques have proven to be crucial for understanding fuel microstructure evolution upon neutron damage accumulation
- EBSD analyses reveal restructuring “in-fieri” at intermediate radial locations
- Commercial fuel characterization is used to evaluate microstructural changes, but deconvolution of effects is difficult
- Separate-effect testing using ion-implantation is a unique tool to study single-parameter effects
- Grain subdivision observed independently of bubble formation. Role of electronic energy losses in UO_2 grain subdivision?

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