Nuclear Materials Mesoscale Possibilities Using Synchrotrons

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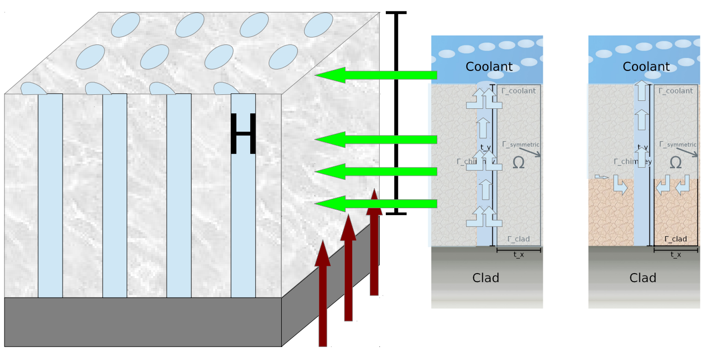
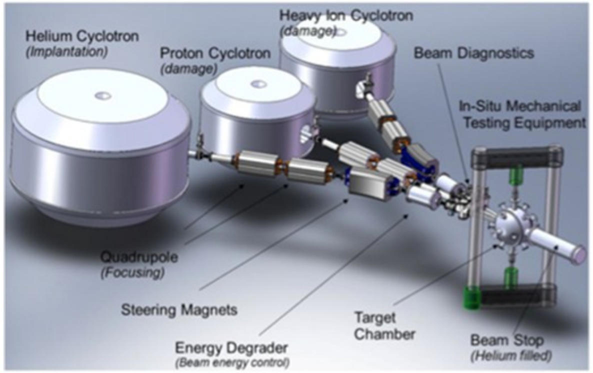
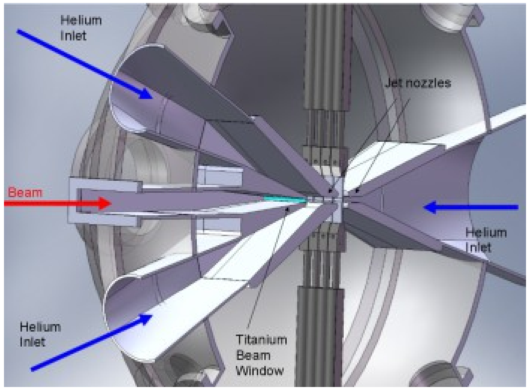
With Thanks To:
CASL (Consortium for Advanced Simulation of LWRs)
Ron Ballinger, Harold Barnard, Ju Li, Dennis Whyte (MIT)
Lynne Ecker, Bill Horak (BNL)







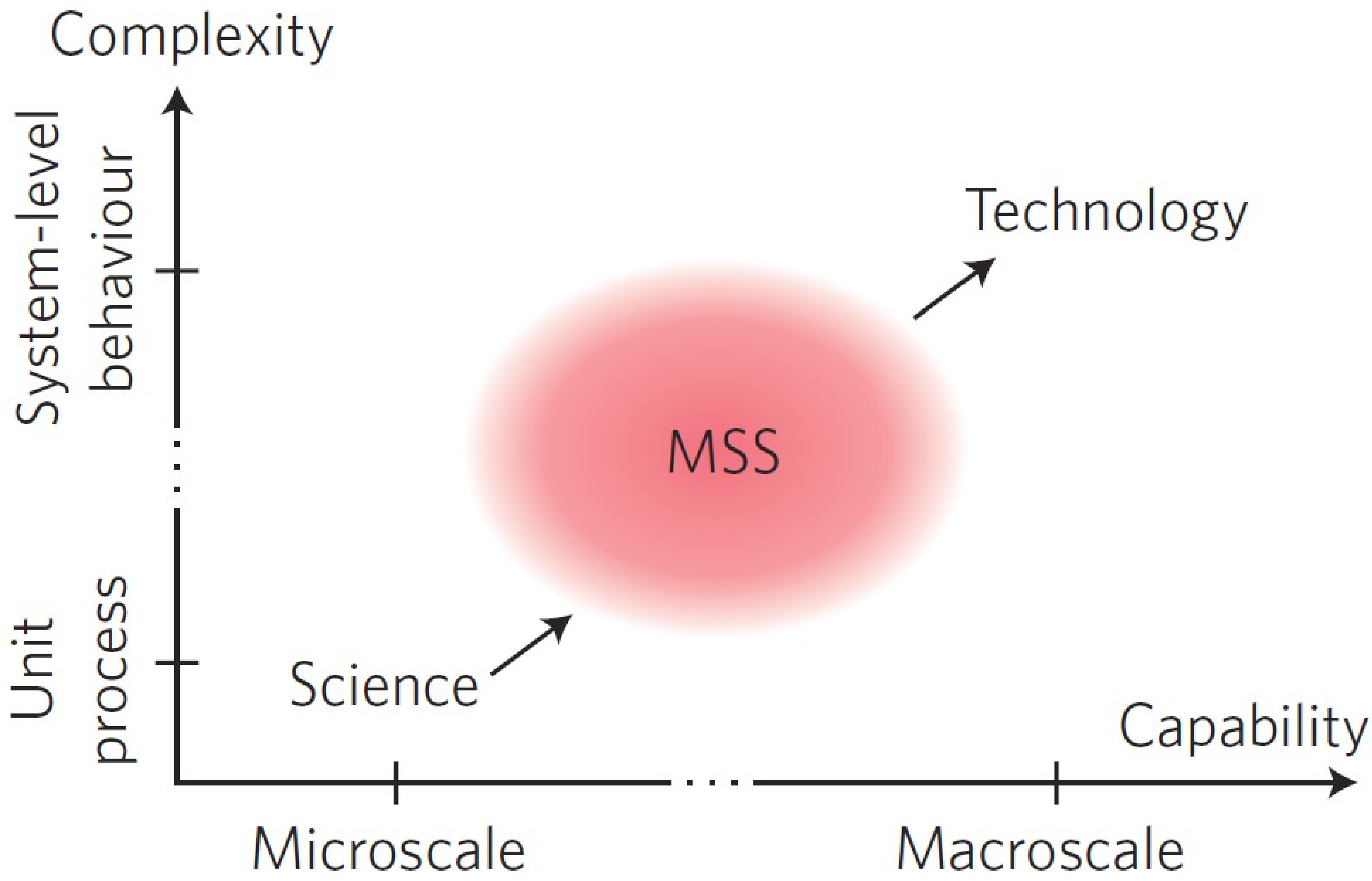
NSLS-II Workshop, Brookhaven National Laboratory, 2013-08-12

Outline

- **Nuclear Materials at the Mesoscale**
- **CRUD[1] and Boiling in Porous Media (no radiation)**
- **Ideal Nuclear Materials Experimental Capabilities**
- **Examples: Void Swelling, Radiation Induced Segregation**

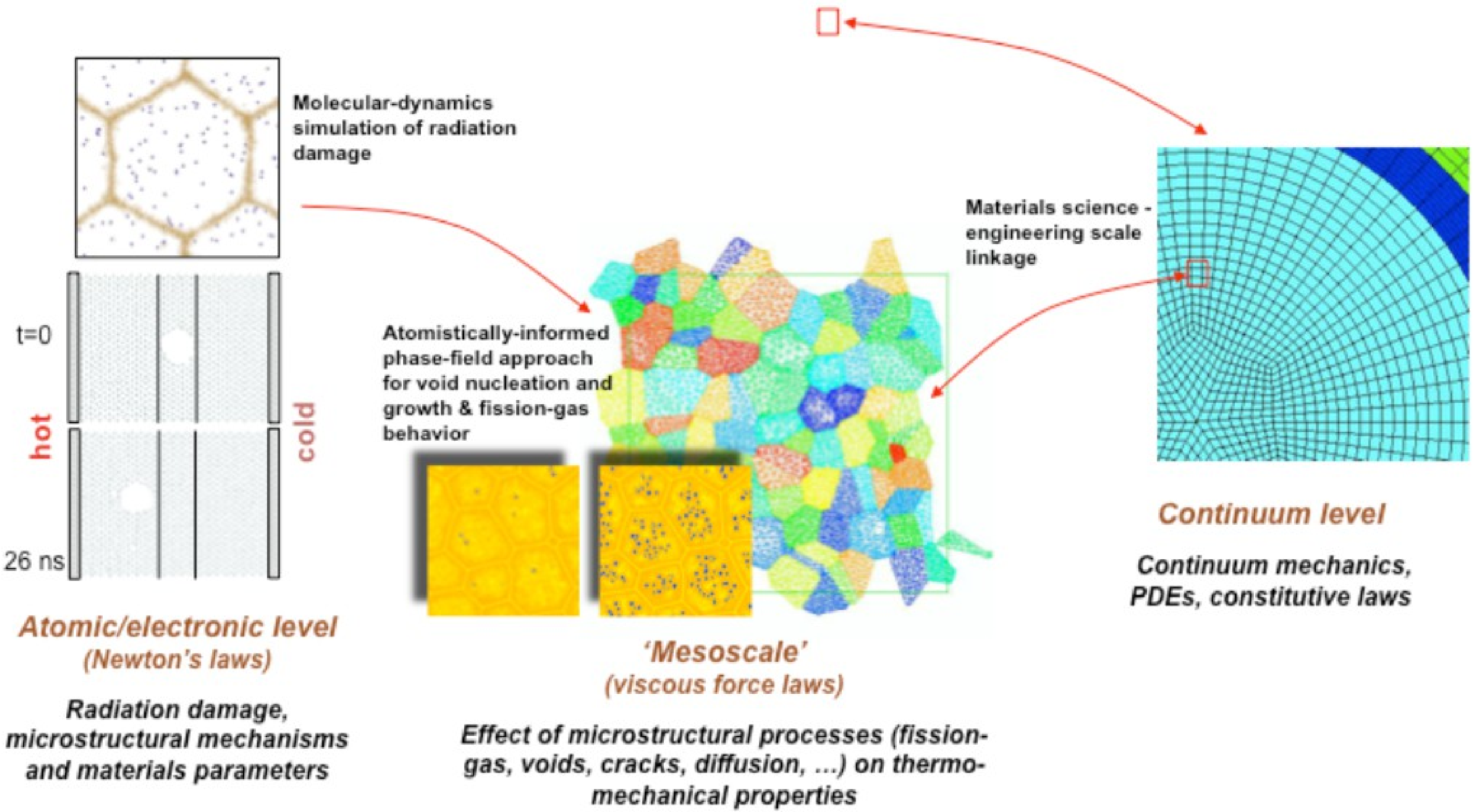
[1]Chalk River Unidentified Deposits

Science Advance vs. Technological Impact: Mesoscale



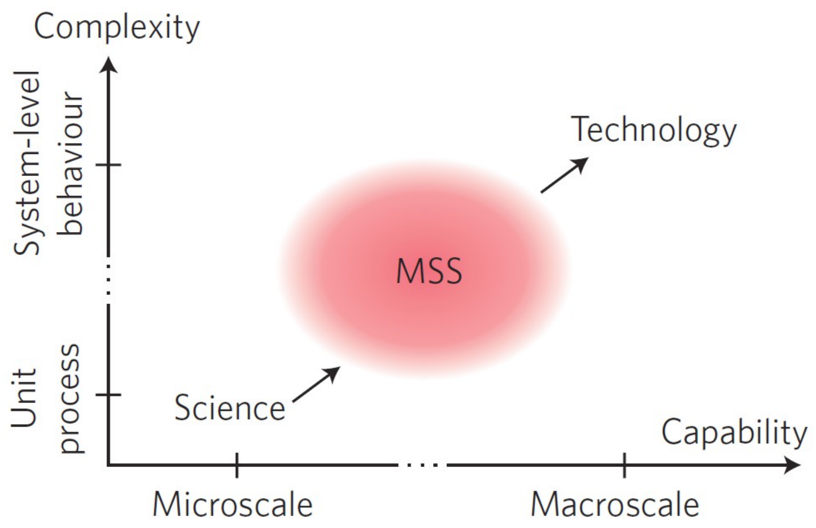
S. Yip, M. P. Short. "Multiscale Materials Modeling at the Mesoscale." Nat. Mater. (In Press), **12**:774-777 (2013).

The Mesoscale: Where It All Comes Together



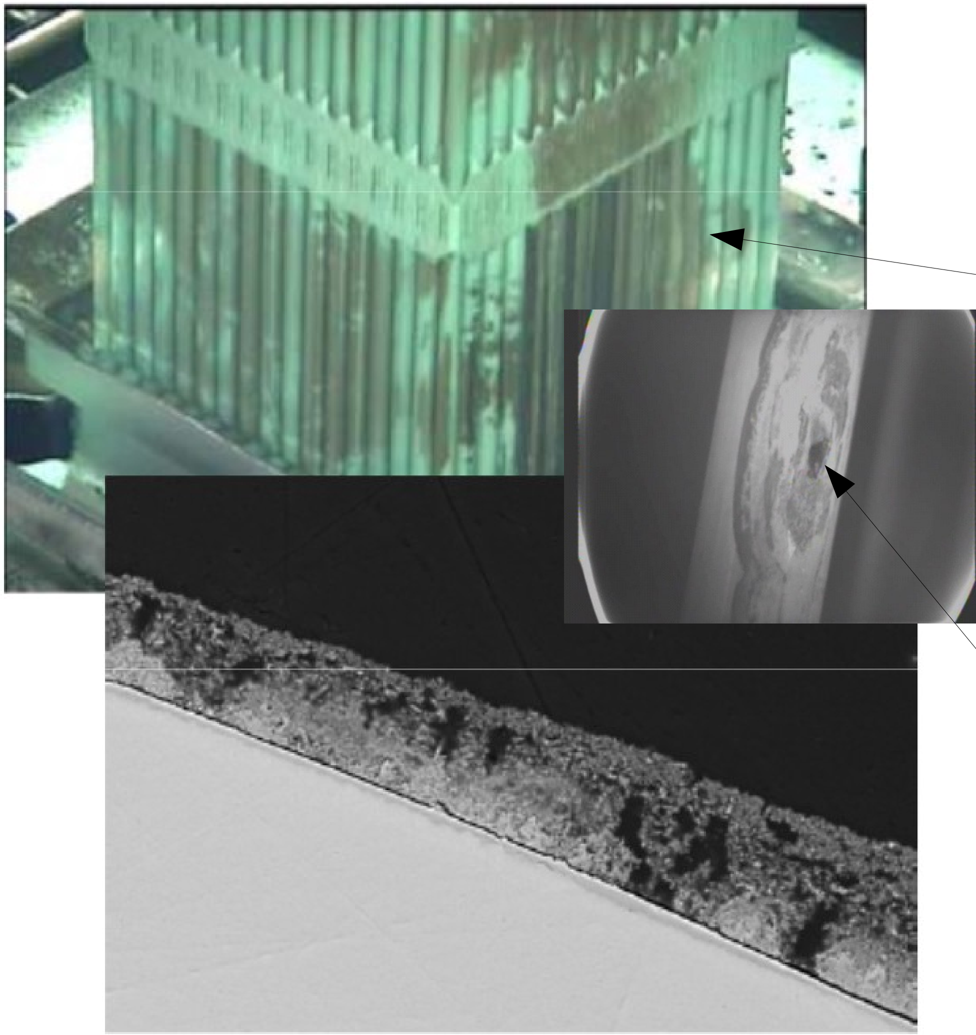
"Report of the Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems." Argonne National Laboratory (ANL), Advanced Photon Source (APS), p. 20, April 2010.

Mesoscale in Nuclear Materials



- Examples
  - Accumulations of radiation defects, changes in mechanical properties
    - Change in ductility
    - Sensitization to corrosion
  - Corrosion / deposition
- Mesoscale effects feel the atomic scale
- Power plants feel mesoscale effects

CRUD: Porous Corrosion Deposition



Images: D. Wells.EPRI P-TAC Summer Meeting, Atlanta, GA, 8/29/2012.

Problem Definition

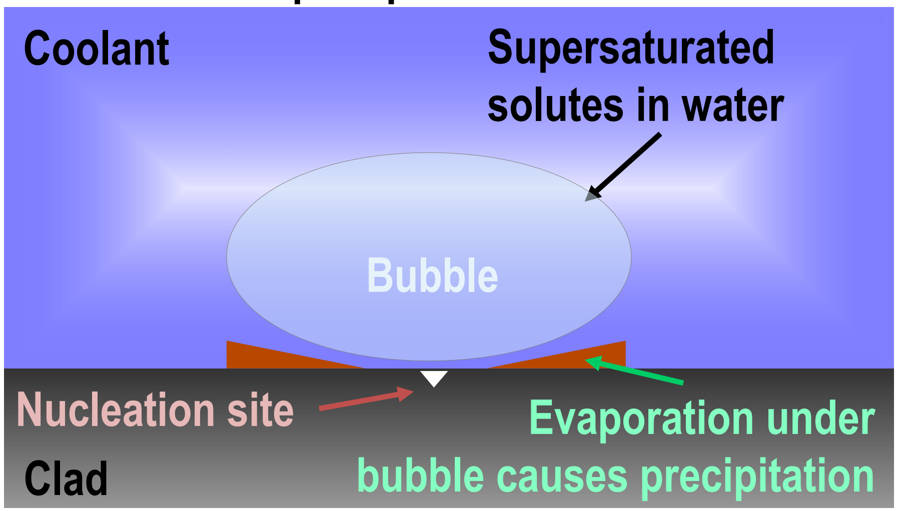
- Reactor internals corrode, releasing products into coolant
- Sub-cooled boiling creates porous deposits
- Boric acid in coolant hides out in CRUD pores

Effects on Nuclear Plant

- CRUD-Induced Power Shift (CIPS) due to boron
- CRUD-Induced Localized Corrosion (CILC) due to degraded heat transfer
- Increased worker dose due to CRUD activation

The Anatomy of CRUD

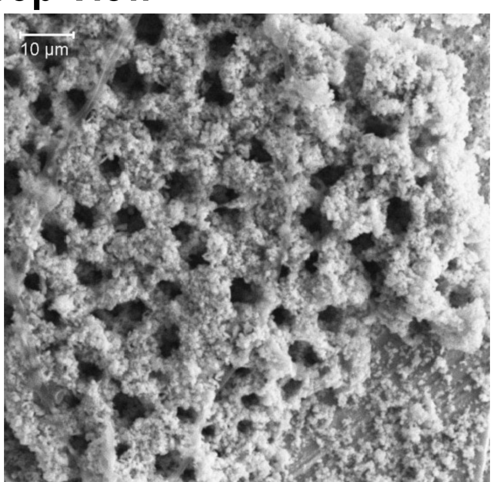
Porous CRUD precipitates underneath bubble



Major Features

- Deposits on upper spans of fuel rods
- Skeleton of oxides from reactor internals
- Boiling chimneys from bubbles
- Typically 5-100 µm thick
- Solubles (Li, B) precipitate in the pores

Top View

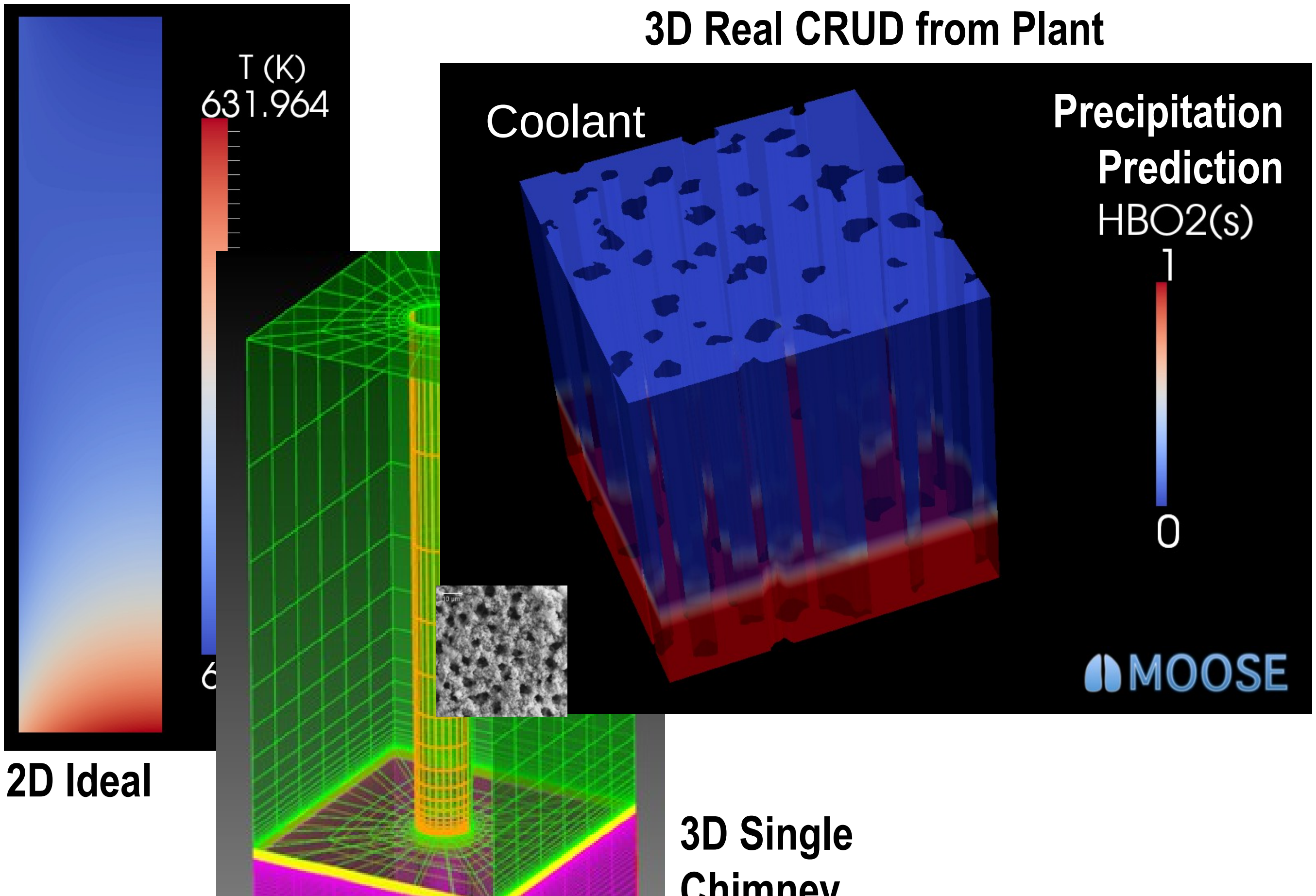


Side View

<u>Images</u>: J. Deshon. "PWR Axial Offset Anomaly (AOA) Guidelines, Rev. 1." EPRI Technical Report #1008102, p. 51 (2004).

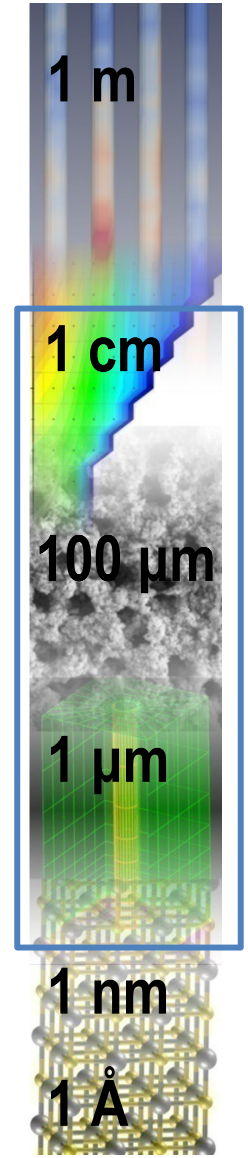
Micro/Mesoscale Model: MAMBA-BDM

3D Real CRUD from Plant



2D Ideal

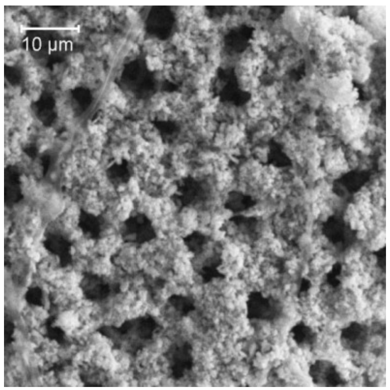
3D Single Chimney



- Written in INL's MOOSE framework, ***geometry/dimension agnostic***, highly parallelizable
- Single chimneys, regular arrays, real CRUD microstructure…

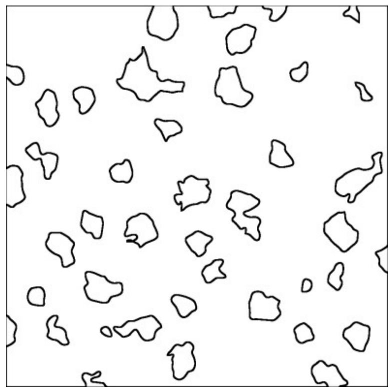
Micro/Meso: Modeling Real 3D CRUD

Acquire Microstructure

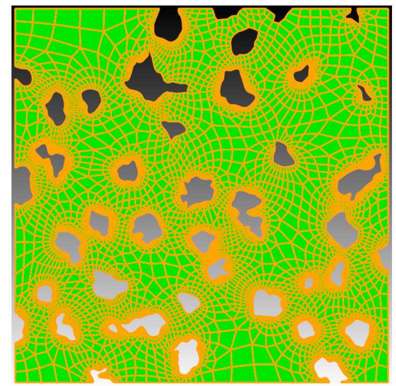


J. Deshon. "PWR Axial Offset Anomaly (AOA) Guidelines, Rev. 1." EPRI Tech. Report #1008102, p. 51 (2004).

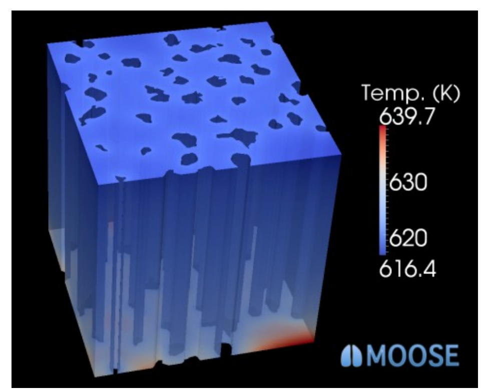
Extract Boundaries



Create CRUD Mesh

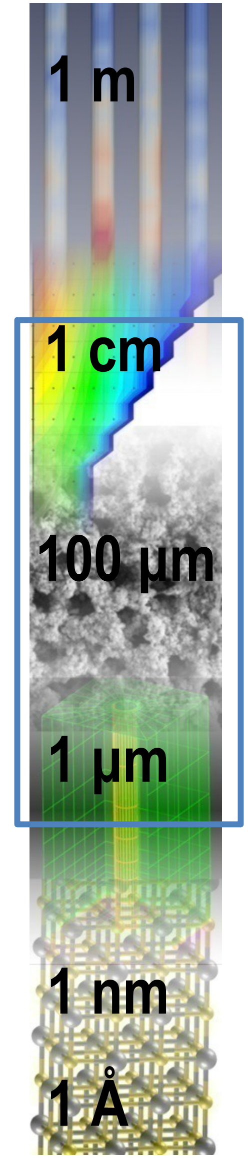


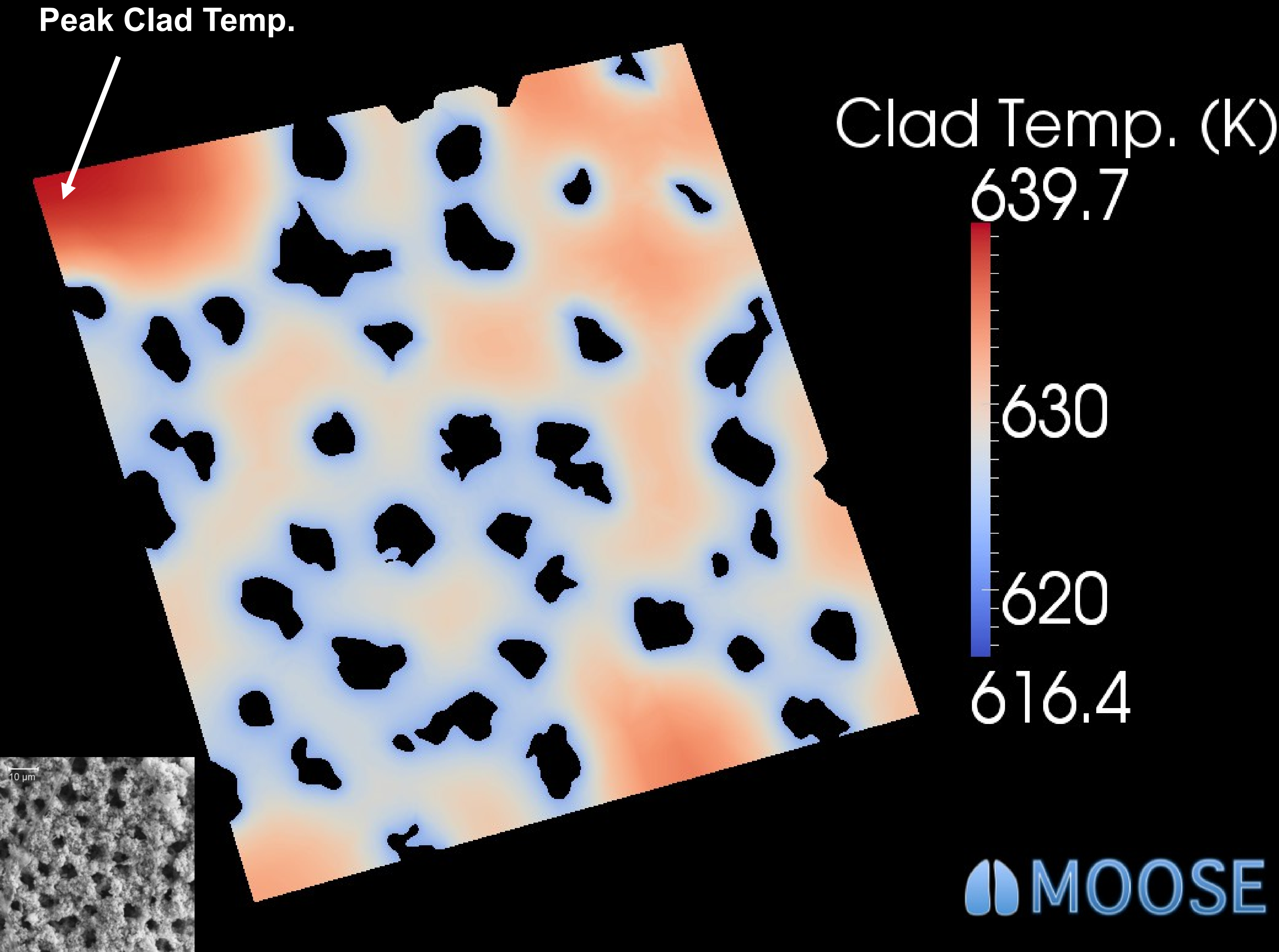
Simulate Real Microstructure



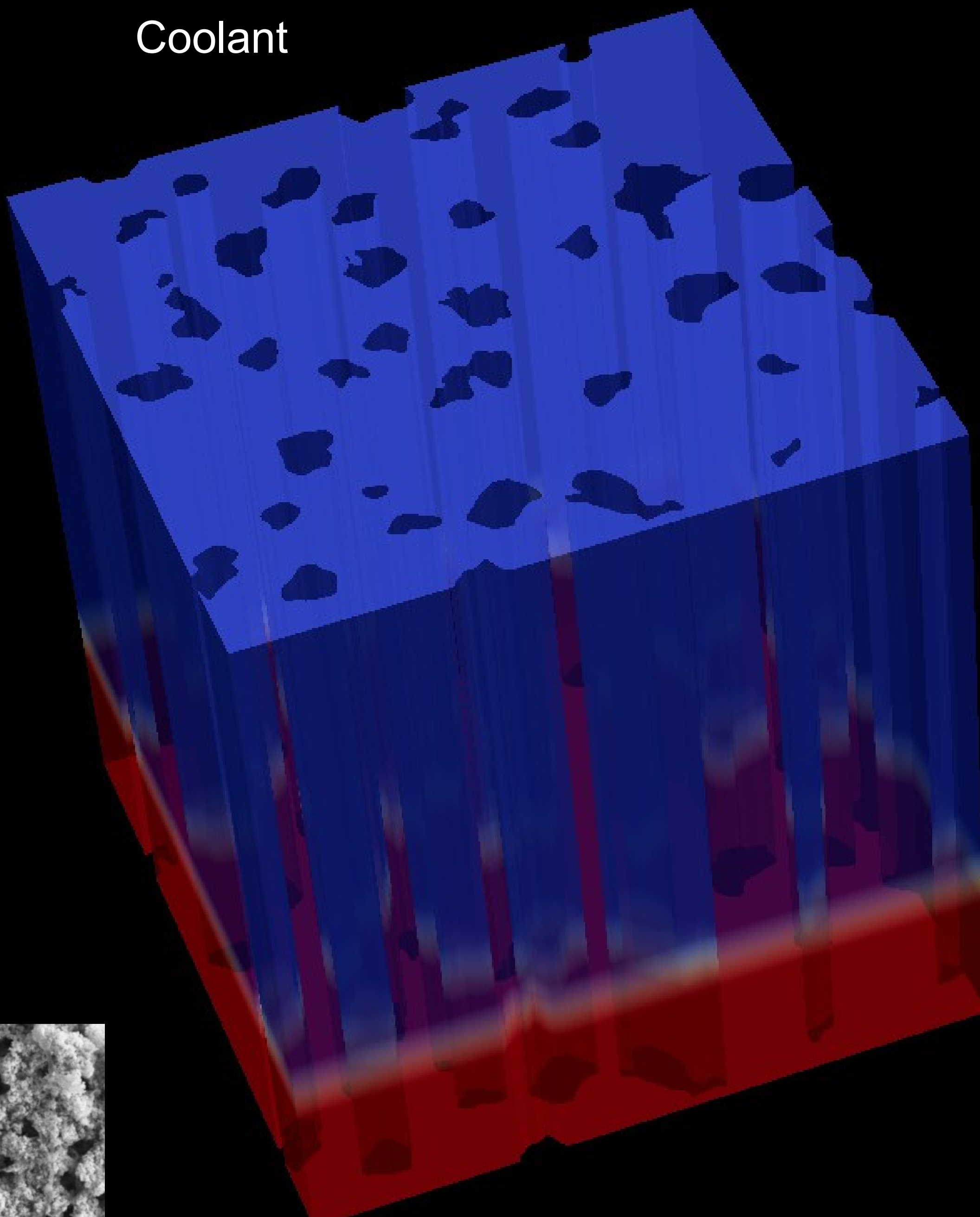
Develop Separate Physical Model

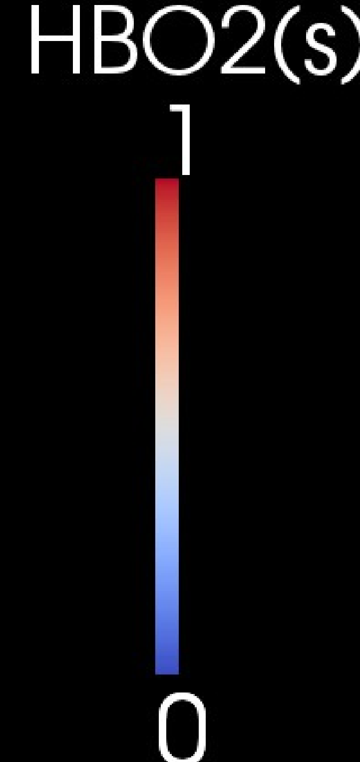
Image: http://www.reptilienzoonockalm.at/Tierbestand.htm

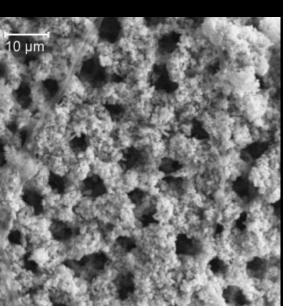



Peak Clad Temp.
Clad Temp. (K)
639.7
630
620
616.4
10 µm

Precipitation Prediction







MOOSE

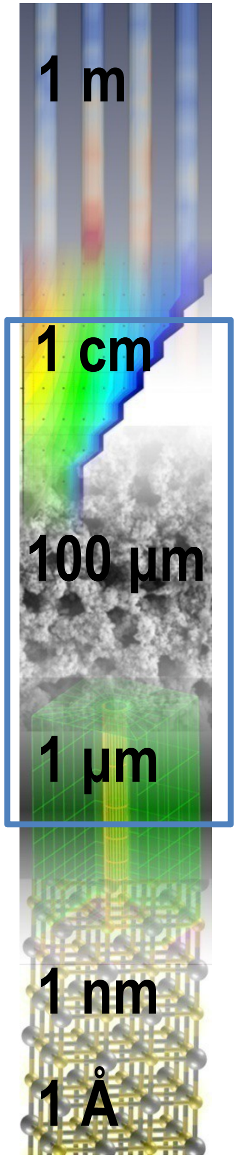
Lack of Validation Shows Missing Mechanism: Boiling & Dryout

- **EPRI Report #1022896**
  - **Measured effective thermal conductivity of synthetic CRUD**
- **Showed very low values compared to MAMBA-BDM**

Ref.	This Work	Models		Experiments	
		[1]	[2]	[3]*	[4]
k_{effc} $\left[\frac{W}{mK}\right]$	5.04	4.40	2.29	0.519 - 1.39	0.82 - 1.82

*Boiling cases (rods 110-117) used

Rod	TC	Crud Thickness (microns)	Porosity (%)	Effective Thermal Conductivity (Btu/hr-ft-°F)
110	1	66	72	0.4130
110	X2	43	76	0.4150
110	X3	88	72	0.5920
111	1	42	73	0.3000
111	2	49	70	0.3450
111	3	46	69	0.3120
111	4	42	56	0.3110
112	1	61	49	NA
112	2	85	66	NA
112	3	58	63	0.3840
112	4	69	67	NA
116	1	99	57	NA
116	2	66	66	NA
116	3	78	62	0.8030
116	4	54	67	0.5870
117	1	120	66	NA
117	2	76	60	NA
117	3	67	59	0.4750
117	4	70	61	0.4790

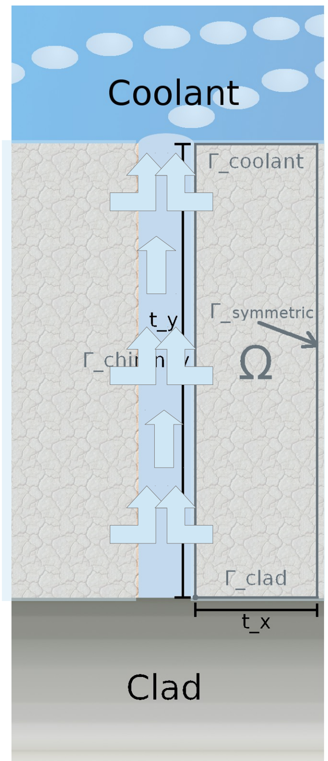


[1] C. Pan, B. G. Jones, and A. J. Machiels. *Nucl. Eng. Des.*, 99:317–327, (1987).
[2] I. Haq et al. *Nucl. Eng. Des.*, 241(1):155–162 (2011).
[4] J. L. Uhle. Boiling Heat Transfer Characteristics of Steam Generator U-Tube Fouling. Ph.D. Thesis, MIT, 1996.

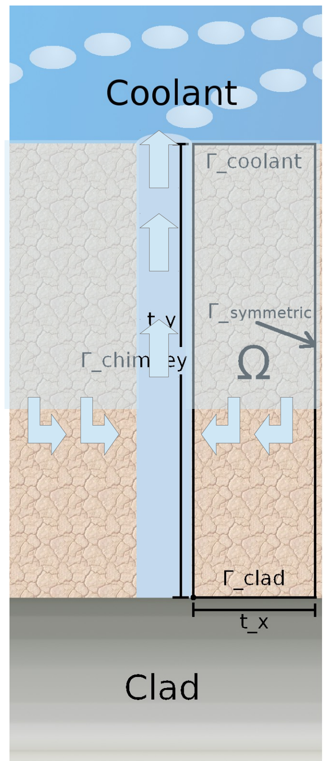
[3] J. Deshon. Simulated fuel crud thermal conductivity measurements under pressurized water reactor conditions. Technical Report 1022896, EPRI, October 2011.

- **Comparison of effective thermal conductivity**
- **MAMBA-BDM overestimates compared to data**
- **<u>Two-phase fluid would correct this, more realistic</u>**

Unknowns: Boiling in Porous Media



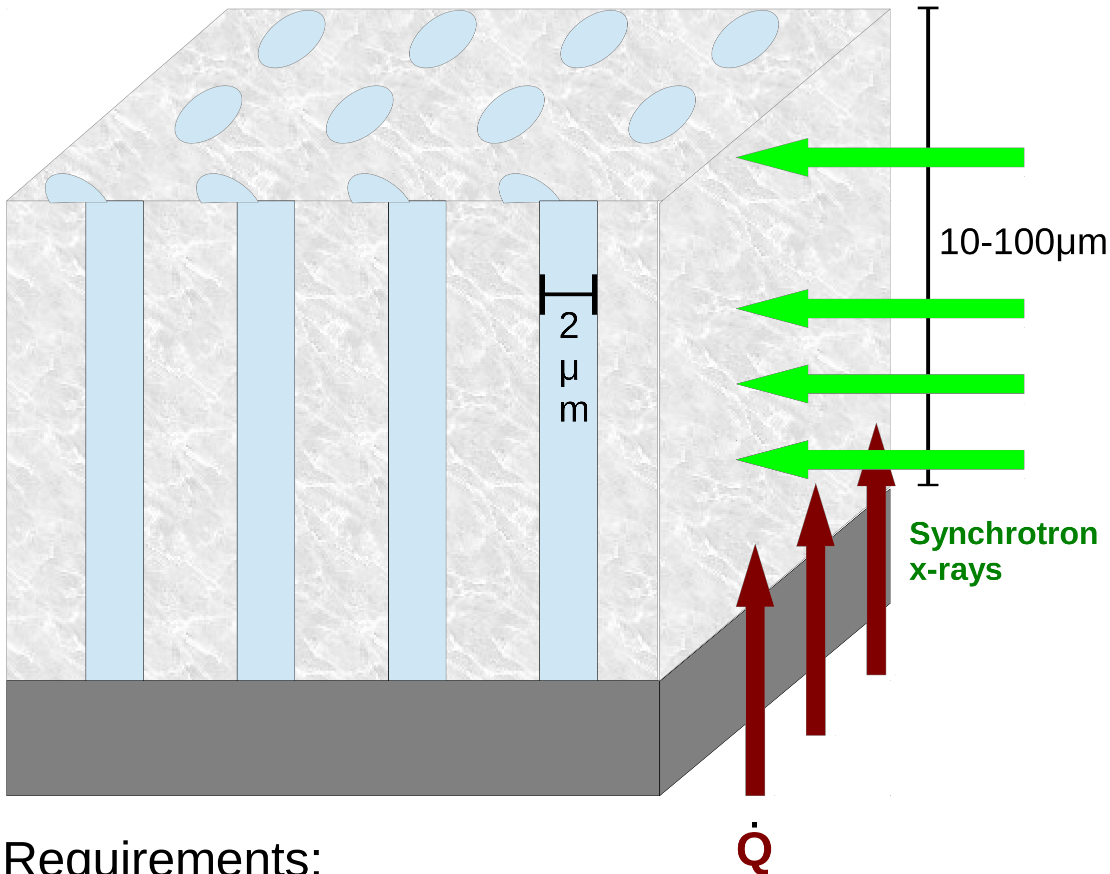
Wick boiling

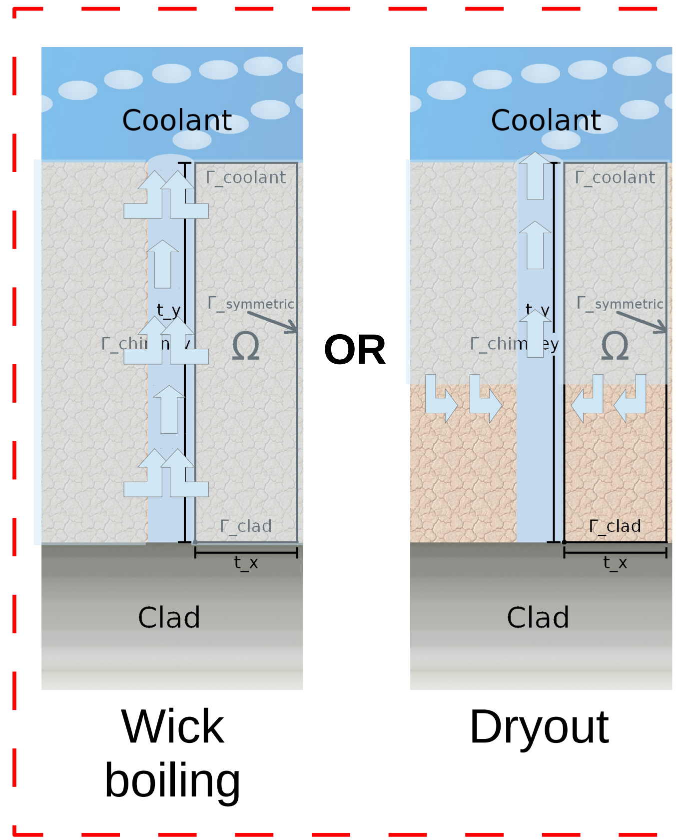


Dryout

- Are “wick boiling” models valid?
  - Most models assume this!
- Is there a dryout region in CRUD?
  - Higher clad temperature, increased corrosion underneath CRUD
  - Few models treat it indirectly
- Extension to other chimney-based porous media (fouling deposits)

Unknowns: Boiling in Porous Media





- Requirements:
  - Boiling experiment at pressure during visualization
  - ~500nm resolution
  - Simulated CRUD, regularly spaced artificial chimneys

Recent Advances in Porous Media with Synchrotrons

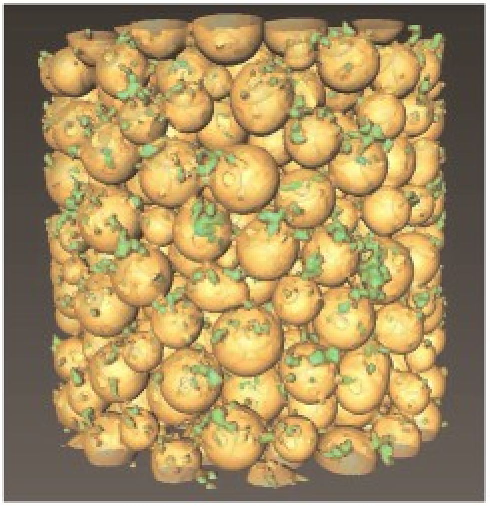


Figure 1. Preliminary three-dimensional X-ray computed microtomography (CMT) biofilm imaging results of biofilm (green) grown in a glass bead pack (gold). The visualization experiment was performed at the Advanced Photon Source (APS), Argonne National Laboratory, using neutrally buoyant, silver-coated hollow glass microspheres (10 μm diameter) as an X-ray contrast agent. The spatial arrangement of the silver particles is interpreted as being attached to the biomass grown within the bead pack.

Iltis et al. "Imaging biofilm architecture within porous media using synchrotron-based X-ray computed tomography." Water Resources Res. **47**:W02601 (2011).

4.5µm

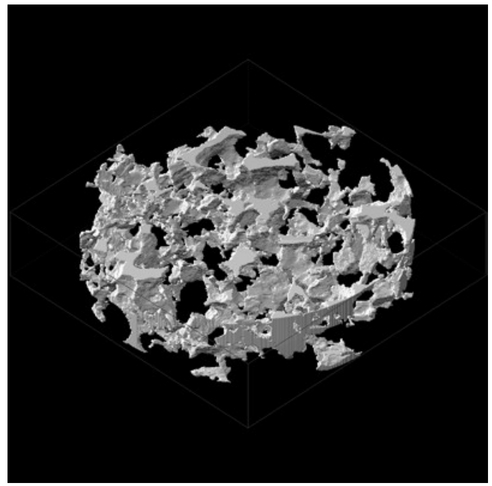
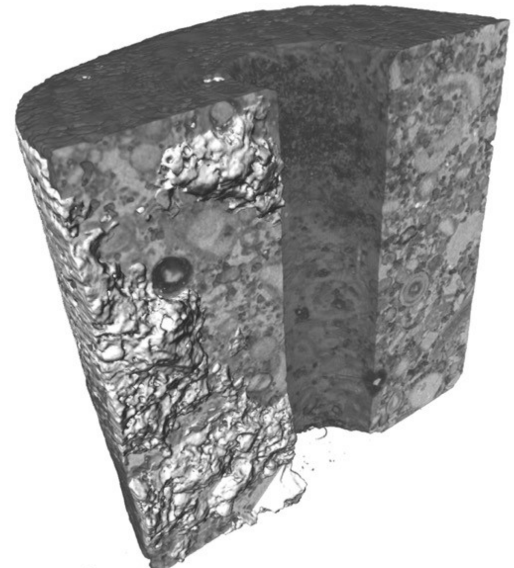


Table 1. Geometric characterization of sample S1a.

500 x 500 x 125 at 1.7 µm Voxels	Total	Cluster	Percolate
Porosity	11.8	11.5	10.1
Surface [m^2]	4.0E-6	3.7E-6	3.2E-6
Specific surface [m^2/g]	1.4E-2	1.3E-2	1.1E-2
Permeability [mD]	360.0	386.2	347.7

F. Enzmann et al. "Synchrotron micro-tomography of porous media for modeling fluid flow." http://www.geochemie.uni-mainz.de/geochemistry/Enzmann.pdf

500x500x125, 1.7µm



b)

F. Renard et al. "3D imaging of fracture propagation using synchrotron X-ray micro-tomography." Earth Plan. Sci. Lett. **286**:285-291 (2009).

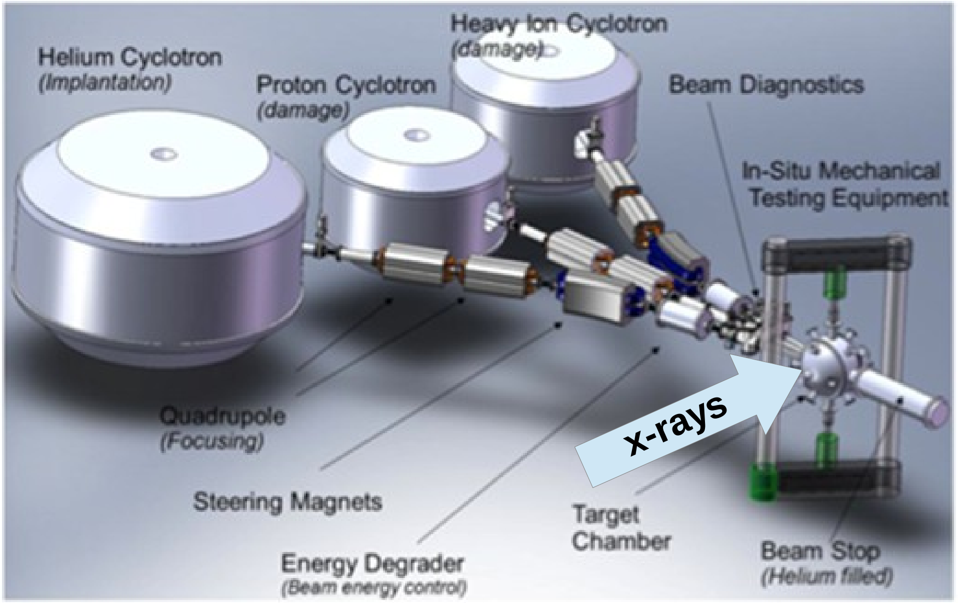
4.91µm

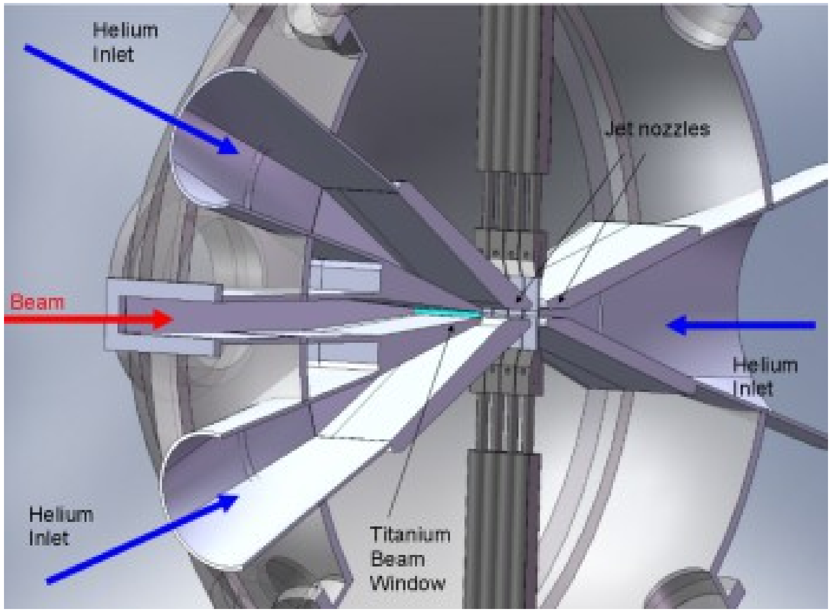
Echoing Previous Statements...

- “Taking apart batteries is something we do because we have to.”
  - *–Esther Takeuchi, one hour ago*
- Now... add radiation, safeguards, and timescales of days-years to the mix!
- In-situ synchrotron visualization can revolutionize this field through *throughput* and *enabling realism*

Ideal Nuclear Materials Beamline w/Radiation Damage

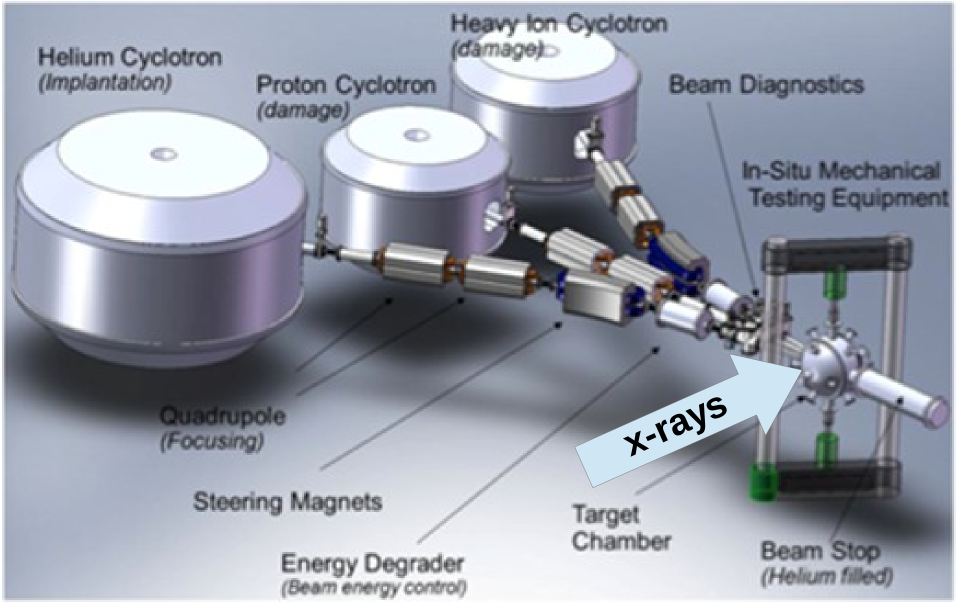
- Triple cyclotron facility with x-ray tomography, phase contrast imaging (for higher contrast), scattering capability
  - Protons (simulates neutrons), helium, heavy ions
- Potential Applications
  - Structural materials, swelling, irradiation creep
  - High dose (fast reactor) materials
  - Flux/energy dependent high-level waste transmutation

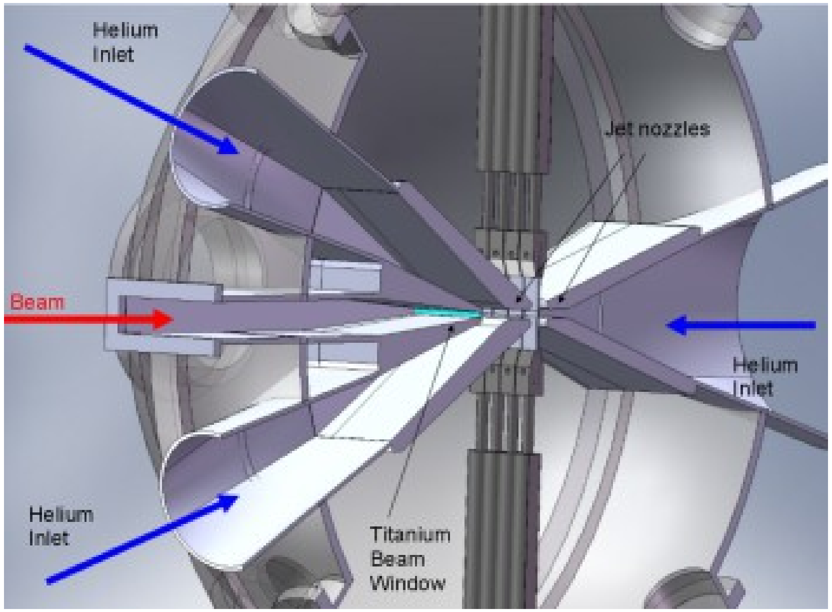




Ideal Nuclear Materials Beamline w/Radiation Damage

- Continuously irradiate sample
- Take x-ray measurements periodically
  - Not feasible with lower flux light sources
  - Not realistic/prototypical with just ion irradiation



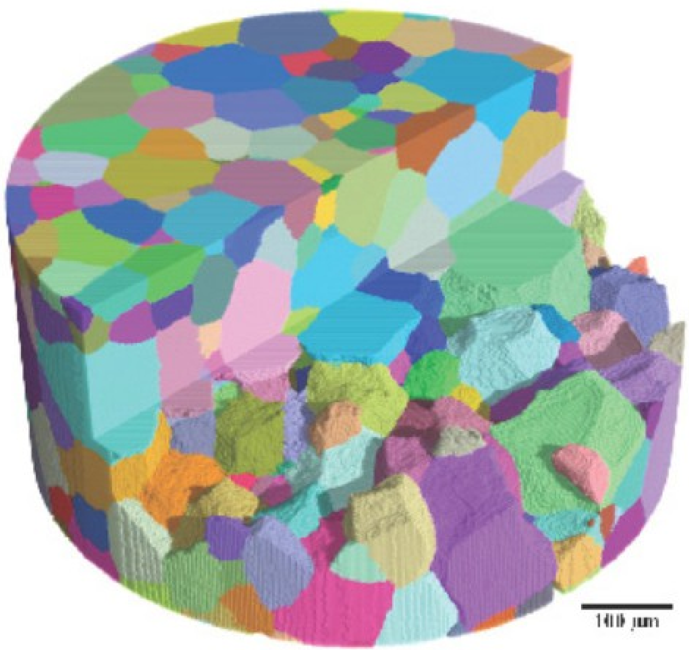


State of the Art (According to Argonne)

In the soft X-ray region, current tomography with resolution on the order of 50 nm is possible [33]. Unfortunately, soft X-rays do not have the penetration depth necessary for studies of either irradiated structure materials or fuels. In the hard X-ray region, sub-100 nm reconstruction is possible [34] at 9 keV with the possibility of pushing the resolution to 15 nm with much lower efficiency. Even at 9 keV, the attenuation length of the X-rays is on the order of 10 μm. This probe depth limitation precludes the study of bulk materials such as clad fuel pellets at these resolutions. The study of bulk irradiated materials requires incident photon energies between 30 and 100 keV, where the attenuation length can extend to 650 μ for spent fuel and as high as 3,400 μ for steels associated with reactor components.

"Report of the Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems." Argonne National Laboratory (ANL), Advanced Photon Source (APS), p. 33, April 2010.

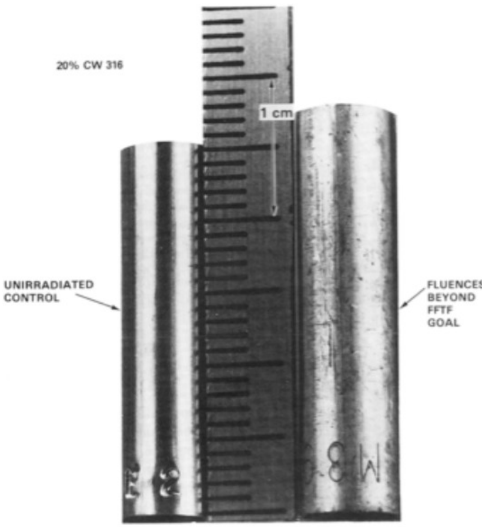
HARD X-RAY IMAGING

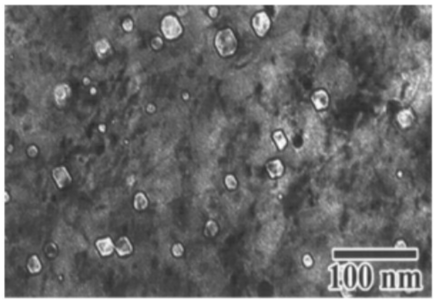


A rendition, obtained by Diffraction Contrast Tomography (DCT), of the three dimensional grain structure of a cylindrical specimen composed of 1008 individual beta-Ti grains. Whereas this image was obtained using 40 keV X-rays, samples such as spent fuel, which contain heavier atoms and are denser, would require even harder X-rays. H. Poulsen et al., Rev. Sci. Instr. 80 (2009) 33905.

Extending to Nuclear Materials

- 5μm resolution possible with good detectors
- Smaller features must be seen
  - Precipitate nucleates
  - Regions of elemental segregation
  - … voids???

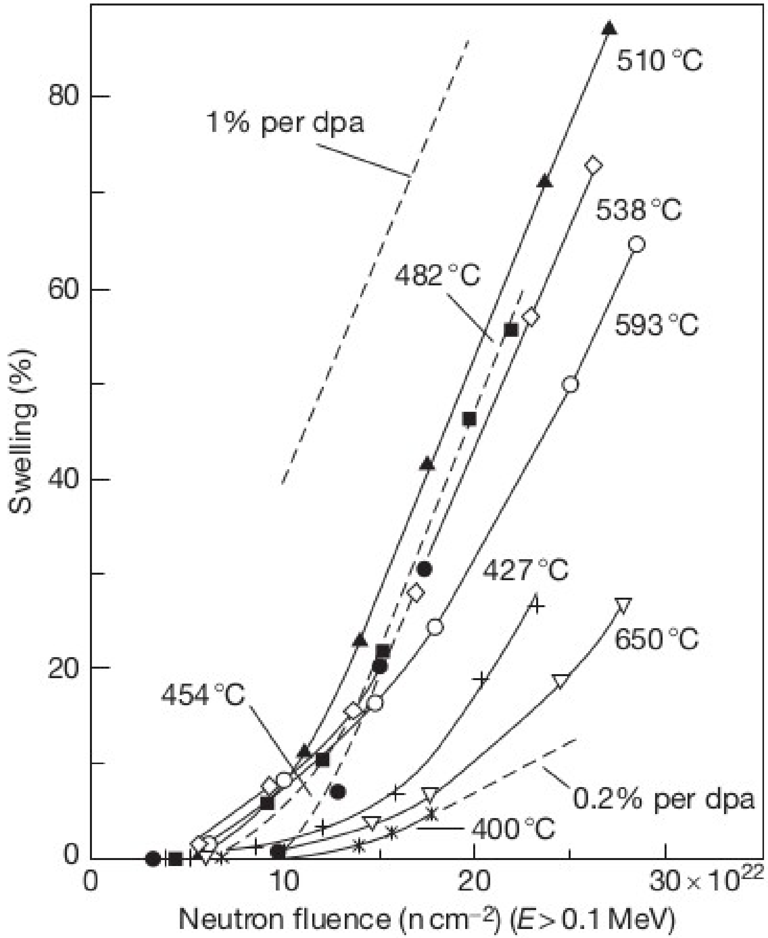




Swelled fuel rods and TEM image of radiation-swelling voids. From *Garner, F., "Irradiation performance of cladding and structural steels in liquid metal Reactors in Materials Science and Technology, A Comprehensive Treatment, Cahn, R., Haasen, P., and Kramer, E., Editors. 1994, VCH: Weinheim*

Example: Predicting Void Swelling

- Upturn in void swelling still not predictable
- Studies done at few doses
- Upturn vs. dose, minor alloying elements not yet observed in-situ

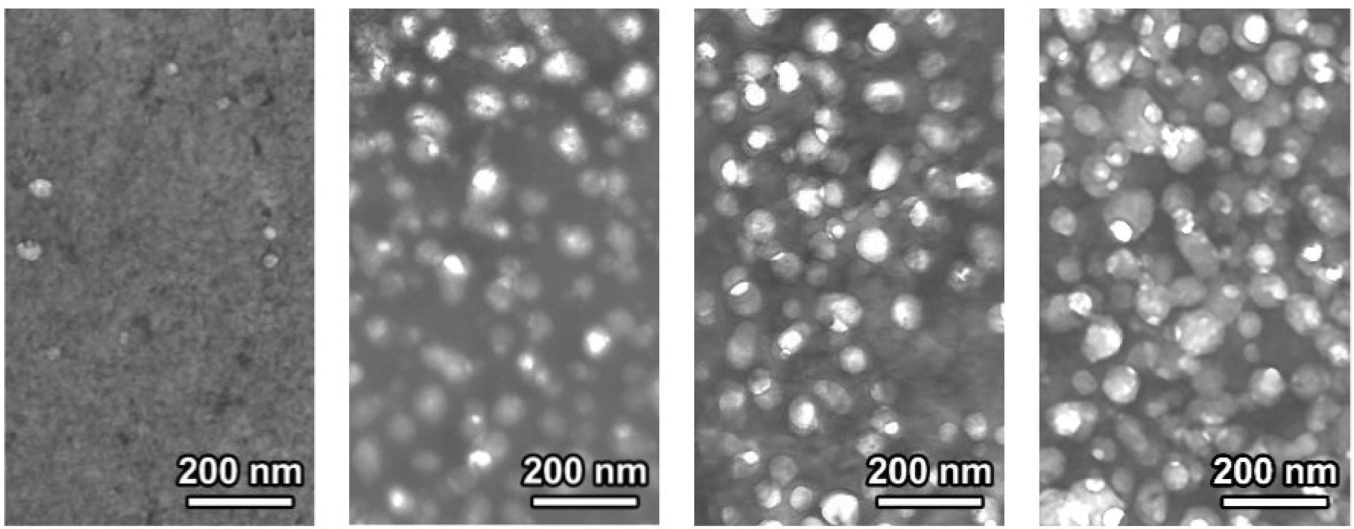


F. A. Garner and D. S. Gelles, in *Proceedings of Symposium of Effects of Radiation on Materials: 14th International Symposium; ASTM STP 1046* (1990).

Void Swelling vs. Dose

Void structure of EP-450 after ion irradiation without He or H at 480°C at ≥150 dpa

150 dpa	200 dpa	250 dpa	300 dpa

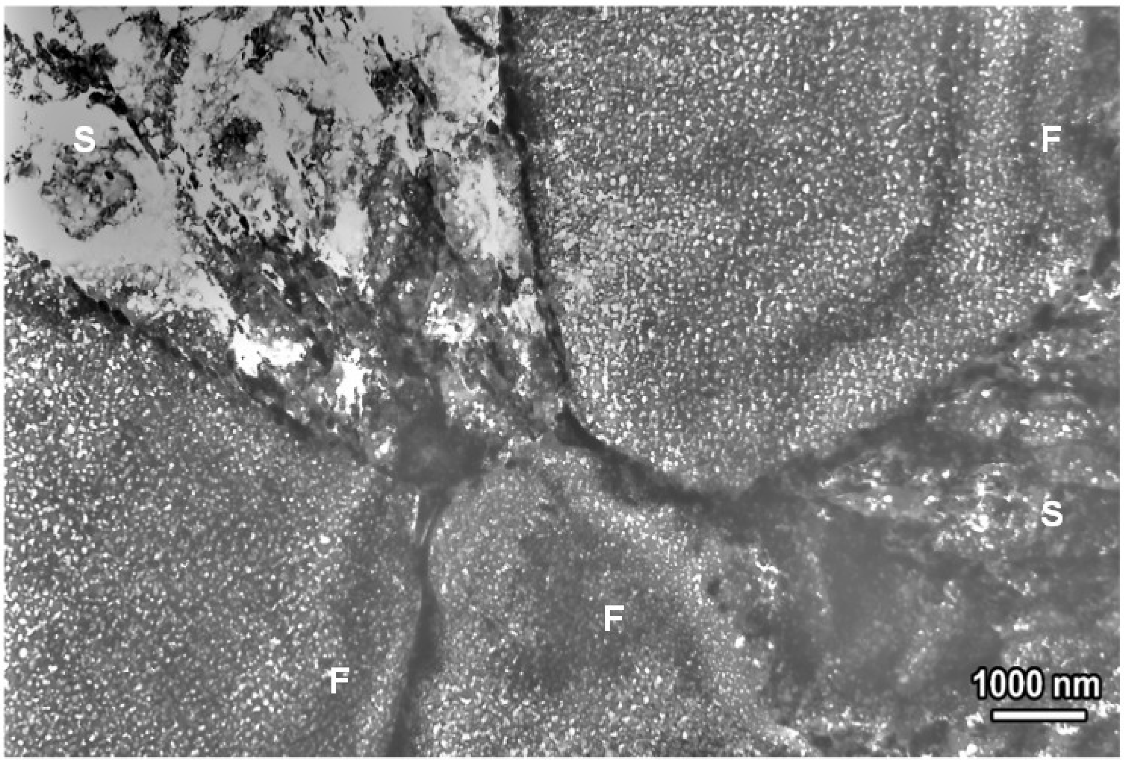


150 dpa	200 dpa	250 dpa	300 dpa
S = 0.6 %	S = 6.9 %	S = 13.3 %	S = 20.4 %
<d> = 28 nm	<d> = 43 nm	<d> = 52 nm	<d> = 58 nm
$\rho = 5.0 \cdot 10^{14}$ cm^{-3}	$\rho = 1.5 \cdot 10^{15}$ cm^{-3}	$\rho = 1.8 \cdot 10^{15}$ cm^{-3}	$\rho = 2.0 \cdot 10^{15}$ cm^{-3}

F. A. Garner et al. "Use of ion irradiation to study void swelling of ferritic and ferritic-martensitic steels at damage levels of 50 to 600 dpa at 400-580°C." Retrieved at http://scatter.nuc.berkeley.edu/files//Cross on August 12, 2013.

Void Swelling vs. Crystal Structure

Structure of EP-450 at 480°C and 300 dpa without gas, showing swelling starts <u>first</u> in ferrite grains



F. A. Garner et al. "Use of ion irradiation to study void swelling of ferritic and ferritic-martensitic steels at damage levels of 50 to 600 dpa at 400-580°C." Retrieved at http://scatter.nuc.berkeley.edu/files//Cross on August 12, 2013.

Another Example: Radiation Induced Segregation (RIS)

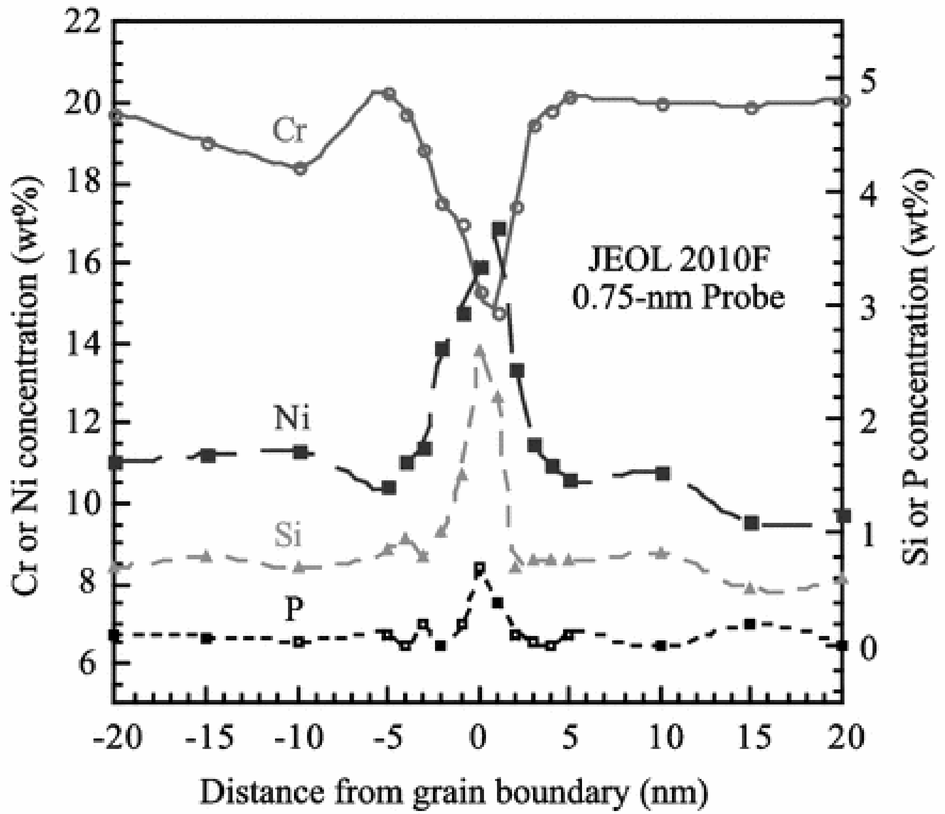


Fig. 6.1. Radiation-induced segregation of Cr, Ni, Si and P at the grain boundary of a 300 series stainless steel irradiated in a light water reactor core to several dpa at ~ 300°C (after [1])

- Too small?

G. Was. "Fundamentals of Radiation Materials Science." Springer, p. 231 (2007).

Questions for the Experts!

Project	Question & Needs
Visualizing time-lapse void swelling	• Can beamline(s) be coupled with ion accelerators? • What radioactive material handling capabilities will exist at NSLS-II? • *What is the best resolution possible?*
Visualizing & measuring radiation induced segregation	• Can elemental segregations around grain boundaries be visualized in phase space? • Can these ideas be tested with in-situ thermal sensitization (like 304SS at 425°C)? • *What is the best resolution possible?*
Determining dryout in porous media	• How will high pressure fluid and solid affect x-ray resolution? • Can high-pressure, high-temperature experiments be placed in the beamline? • *What is the best resolution possible?*