SAND2017-1599PE

Integration of Meso-scale Microstructural Modeling for Engineering Materials Development

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and
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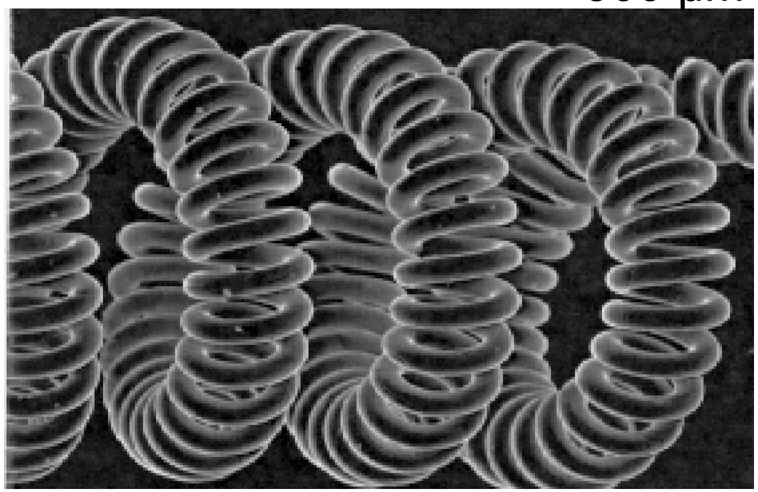
Recrystallization controls the properties of tungsten lamp filaments.

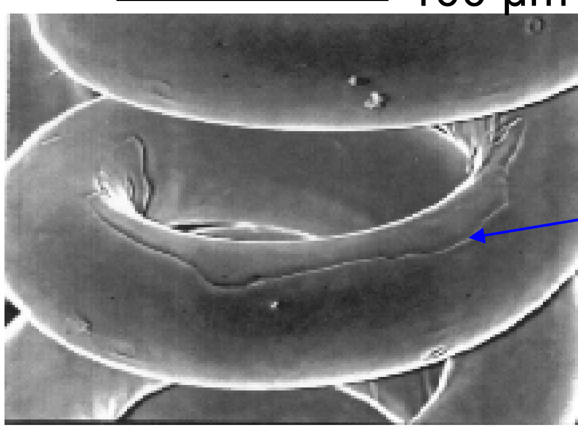
500 µm



- Lamp filaments are a coiled coil of 55 µm diameter tungsten wire
- 40% plastic strain imparted in final coiling
- Filaments operate at 0.8 T_m under shear stresses up to 25 MPa

⇒ Recrystallization occurs during first lamp power up

100 µm



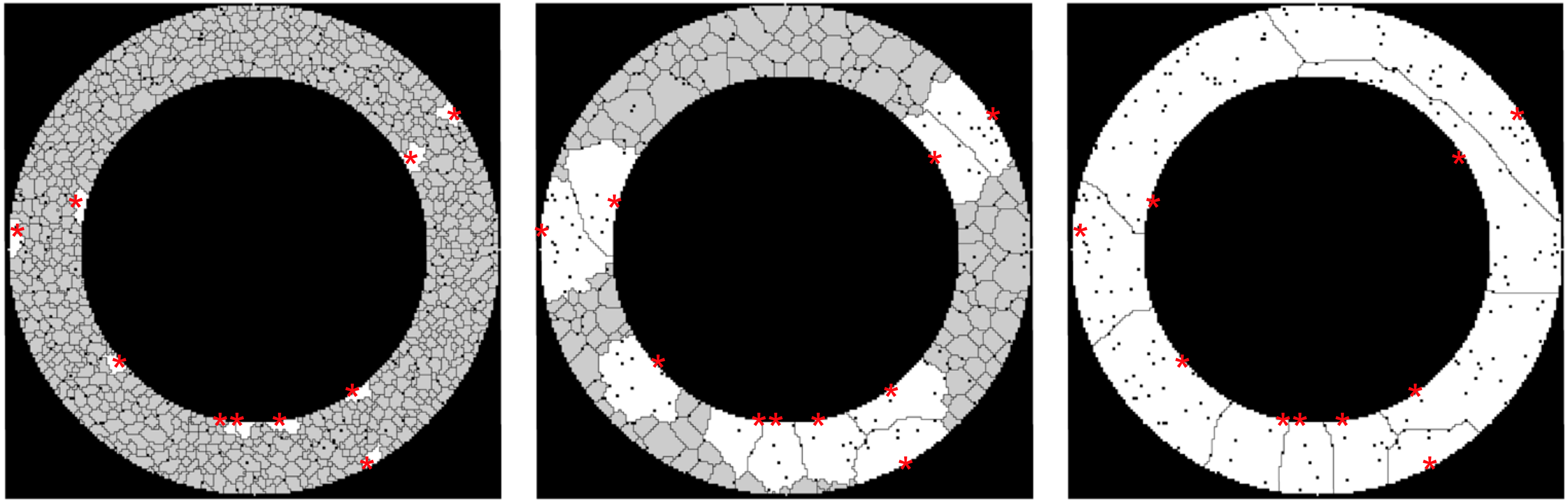
- To minimize creep in service, tortuous boundaries, radial to the filament axis are desired

E.A. Holm, 1996

Micrographs courtesy of John Selverian, Osram-Sylvania

Recrystallization in lamp filaments depends on nucleation parameters

Example: 2-D toroidal specimen, low density of recrystallized nuclei at surface, uniform strain with unrecrystallized $H/\gamma=1$, 0.5 v/o potassium bubbles



t=1000 timesteps   t=10,000   t=100,000

- Recrystallized structure is relatively insensitive to strain energy distribution, pinning particles, prior grain structure, etc.
- Nucleus position, frequency, and growth behavior govern final grain boundary positions.

⇒ However, nucleation parameters are simulation inputs.

E.A. Holm, 1996

What is microstructural, meso-scale modeling?

It is modeling of:

• Grain structure:	Grain size, shape, distributions, … evolution during fabrication and service. Restructuring of FR fuels
• Additional phases, pores, gas bubbles,…	Their size, shape, connectivity, the interfaces between them, their evolution, …
• Defects:	Dislocations, interfaces, stacking faults, twins, … Irradiation damage of vessel, high-burnup rim structure
• Thermodynamics and kinetics:	Energy and interaction between microstructural features leading to complex microstructural evolution.
• Phase-transformation and composition changes:	Component segregation, FP diffusion, formation of precipitates.

Objective and Agenda

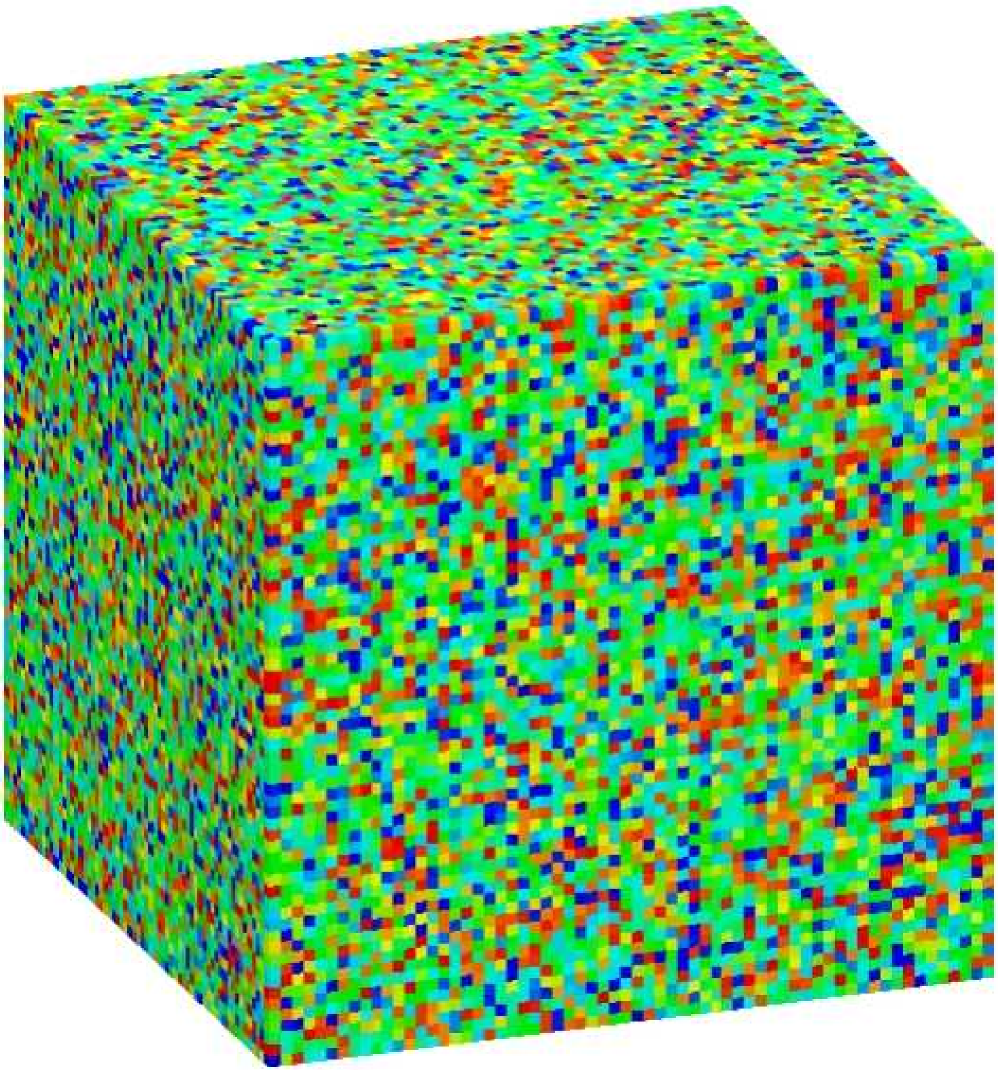
Objective: Describe how meso-scale microstructural simulations can be used to design materials for engineering applications

- **Example of light bulb filaments**

Outline:

- **What are meso-scale models?**
- **Examples of how they can be used**
  - **Grain growth in T-gradient**
  - **Sintering**
  - **Welding**
  - **Fission gas bubbles**
  - **Reorientation of Hydrides**

Potts kMC Simulation of Grain Growth



Grain Growth

•EOS, E = E_{int}

- $$E = \frac{1}{2}\sum_{i}^{N}\sum_{j}^{N} J\left(1-\delta_{ij}\right)$$
- $$P = \exp\left(\frac{-\Delta E}{kT}\right)$$

Shown to simulate

•Curvature-driven grain growth

•Correct kinetics, topology and grain size distribution.

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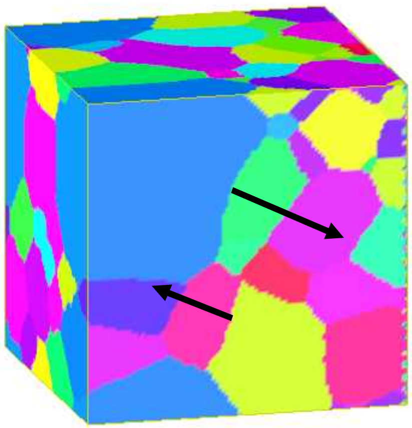
Grain Growth in a Thermal Gradient

Thermal gradient is simulated by changing the mobility of the grain boundaries as a function of temperature.



$$v = -M\kappa$$

$$v = -M_o \exp\left(\frac{-Q}{k_B T}\right)\kappa$$

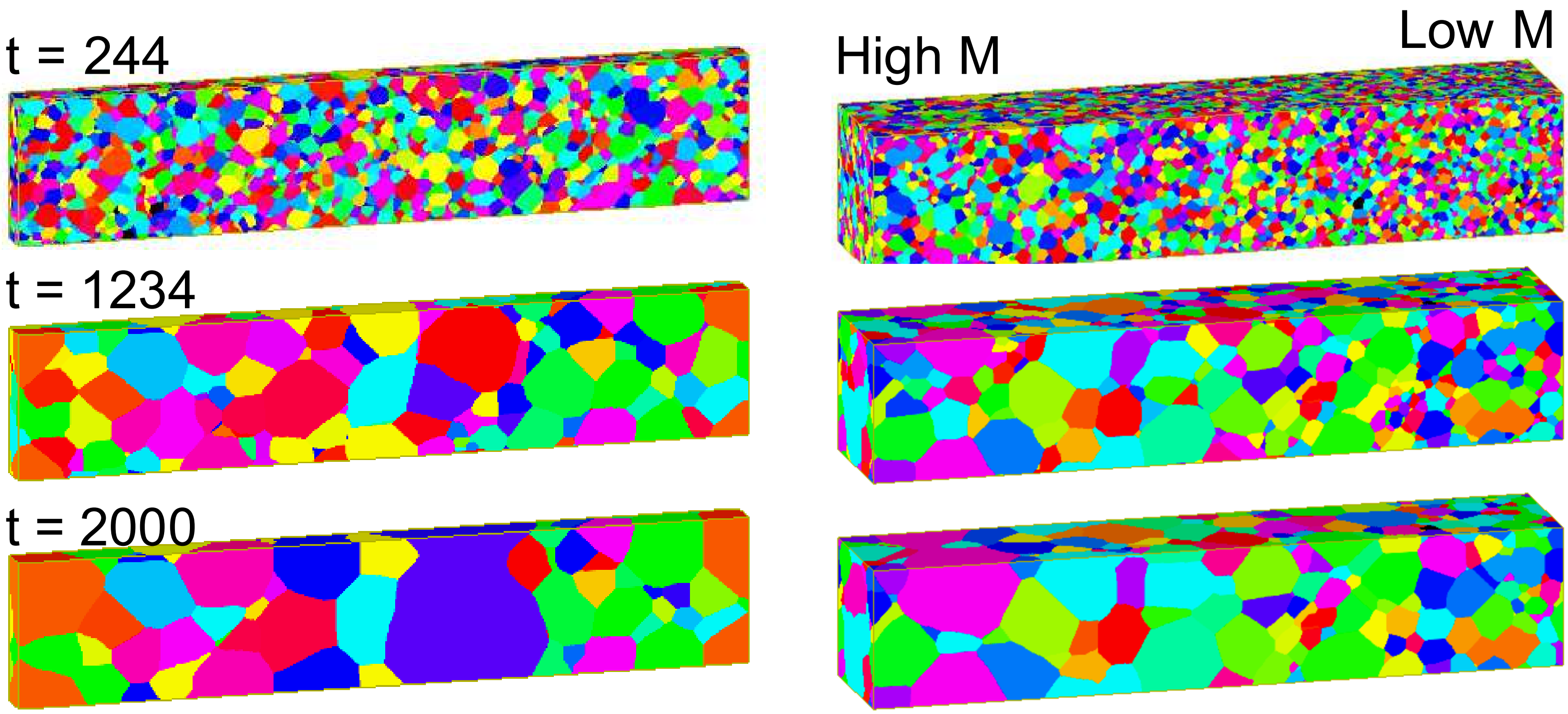
Thermal gradient is implemented in the model by making the probability of grain growth events a function of temperature

$$P = M(T)\exp\left(\frac{-\Delta E}{k_B T_s}\right) \quad \Delta E>0$$

$$P = M(T) \quad \Delta E\leq 0$$

Polycrystalline grain growth in a thermal gradient

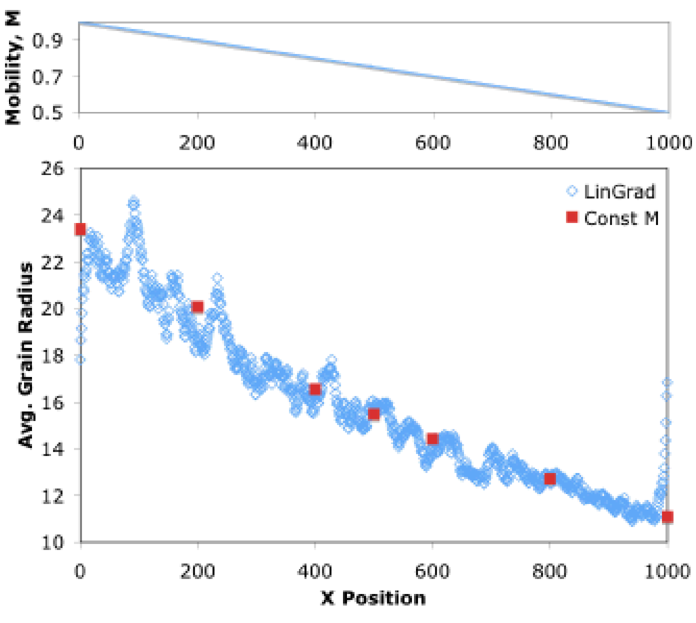
Isothermal grain growth in compared to one with linear mobility gradient

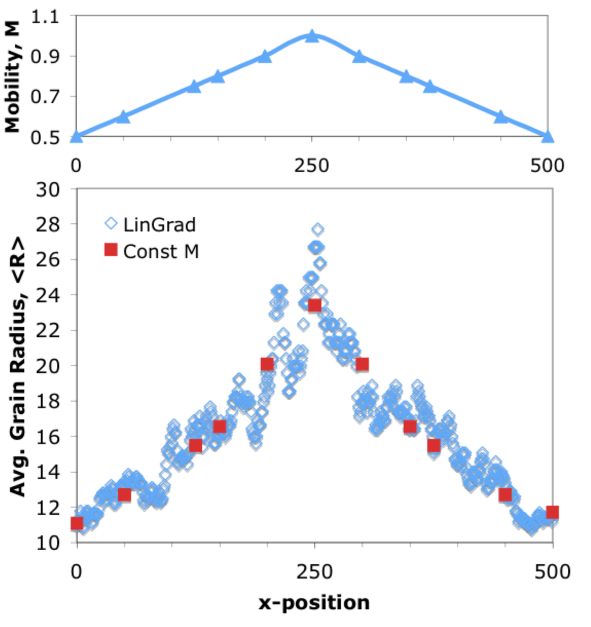


As expected, grains grow faster in the high temperature region where grain boundary mobility is high.

Comparison of grain growth kinetics: isothermal to non-isothermal

Excellent agreement is seen in grain size between simulations with thermal gradients and isothermal at the local temperature when the thermal gradient is gradual, but where there is a sharp discontinuity, contiguity requires homogenizing the grain size.





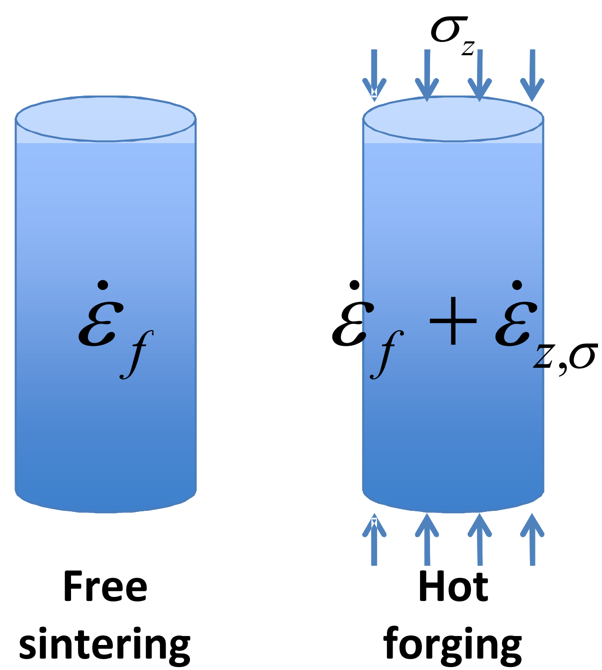
In smoothly varying thermal gradients, the kinetics of grain growth are locally normal.

Experimental Measurement of Sintering Stress

- The usual method for determining sintering stress is loading dilatometry

$$\dot{\varepsilon}_f = \frac{P_L}{3K}$$

$$\dot{\varepsilon}_z = \dot{\varepsilon}_f + \frac{\sigma_z}{3(1-2\upsilon_p)}$$



- Pre-sinter powder compacts to different densities
- Measure the free sintering strain rate, $\dot{\varepsilon}_f$ and the loaded strain rate, $\dot{\varepsilon}_z$
- Calculate the sintering stress, P_L

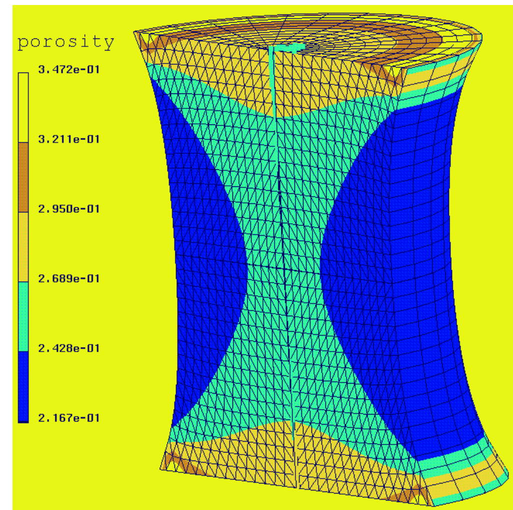
Zuo, Aulbach & Rodel, Acta Met, 2003

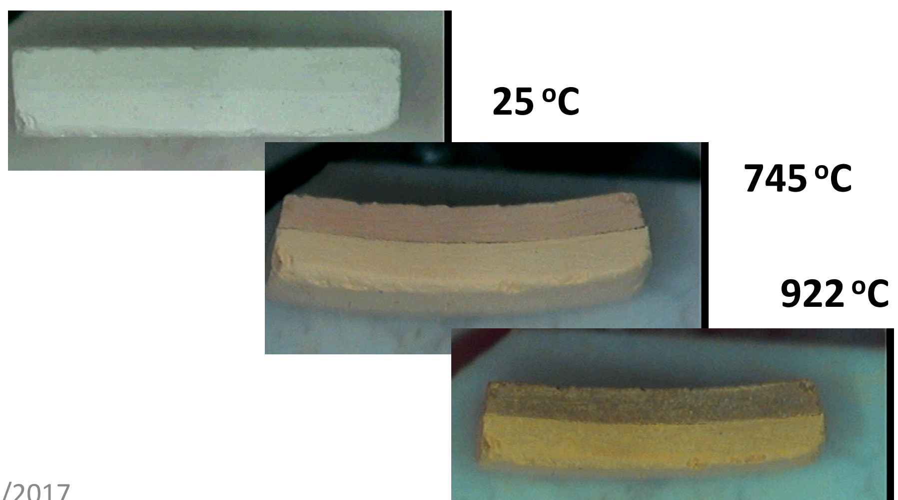
Sintering Stress Can Be Used To Tailor Multi-Materials Ceramics

Simulate constrained sintering with the accompanying shape distortion

- Variations in density
- Multi-layered materials
- Functionally graded materials
- Powder packing defects

At the continuum, component scale.

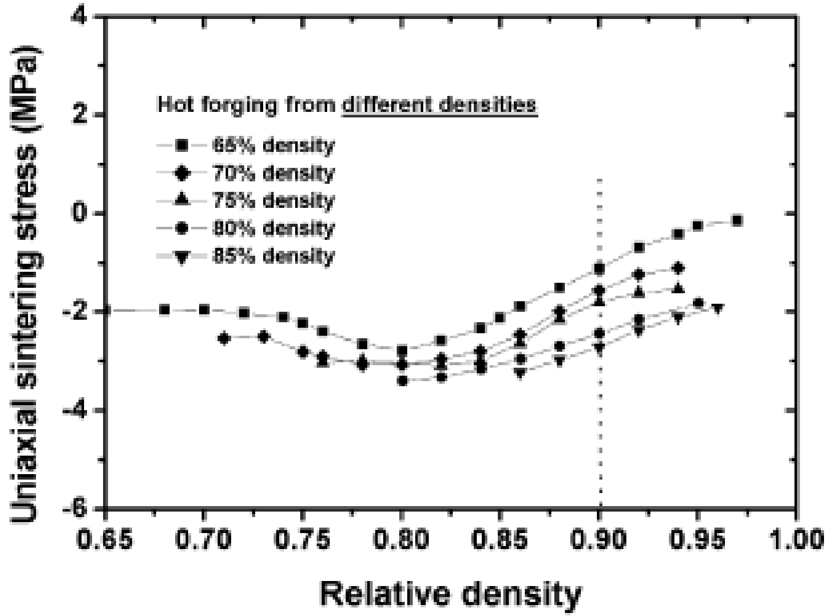


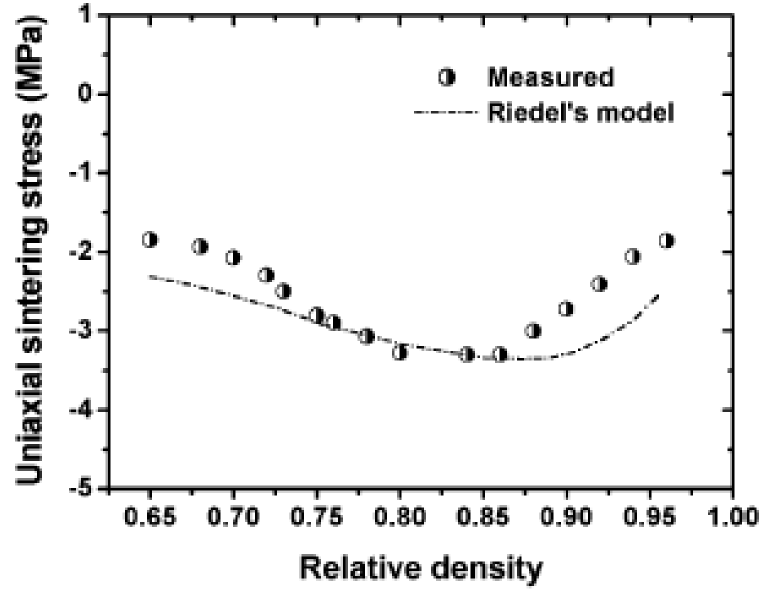


Experiments by T. Garino

Experimental Measurement of Sintering Stress

- **The usual method for determining sintering stress is loading dilatometry**





- **Assumes microstructural evolution during hot forging & free sintering are the same**
- **Zuo et al. showed microstructural evolution is not the same for these conditions and many instantaneous measurements must be made.**

Zuo, Aulbach & Rodel, Acta Met, 2003

Sintering Stress

- **Driving force, ΔG, is the total change in free energy during sintering**
  - **In response to grain boundary energy γ_{gb} pore surface energy γ_s contributions**

$$E = A_{gb}\gamma_{gb} + A_s\gamma_s$$

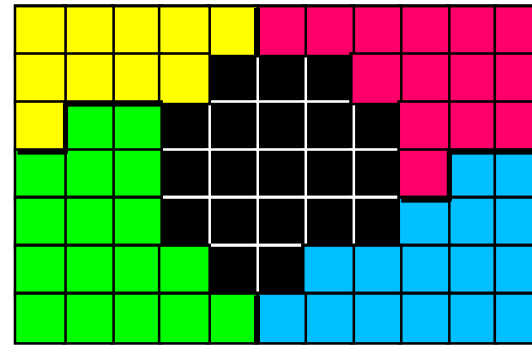
- **Sintering stress, P_L, is the inherent sintering stress due to capillarity for densification.**

- **Energy Method** $$P_L = \frac{E_s(V_o + \Delta V) - E_s(V_o)}{\Delta V} = \frac{\partial E_s}{\partial V}$$

- **Curvature Method** $$P_L = \gamma_s \overline{H} = \gamma_s \frac{\frac{1}{2}\iint_S \left(\frac{1}{r_1} + \frac{1}{r_2}\right) dS}{\iint_S dS}$$

- **Force balance** $$P_L = \sum F_N$$

Sintering Stress
Measuring from Simulations



Energy Method

$$E = \frac{1}{2}\sum_{i=1}^{P}\sum_{j=1}^{26} J_S\left(1-\delta\left(q_i, q_j\right)\right)$$ **For all pore sites q_i**

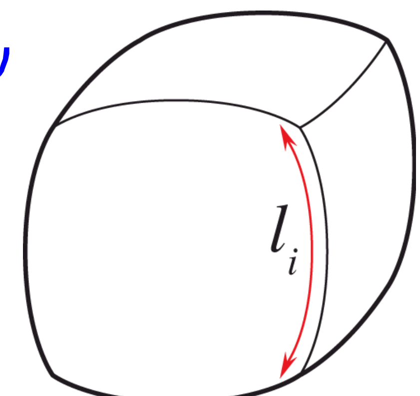
Curvature Method

- **For a polyhedron**
  - **Integral mean curvature**

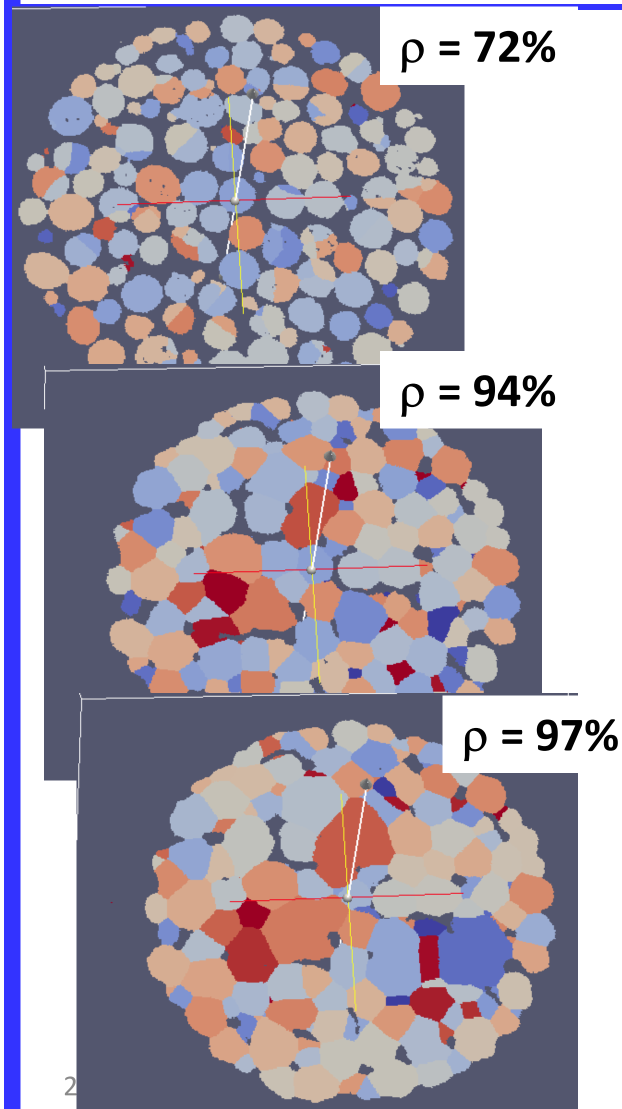
$$M_v = \iint_S \frac{1}{2}\left(\frac{1}{r_1}+\frac{1}{r_2}\right)dS + \frac{1}{2\pi}\sum_{e=1}^{E} l_e \beta_e$$

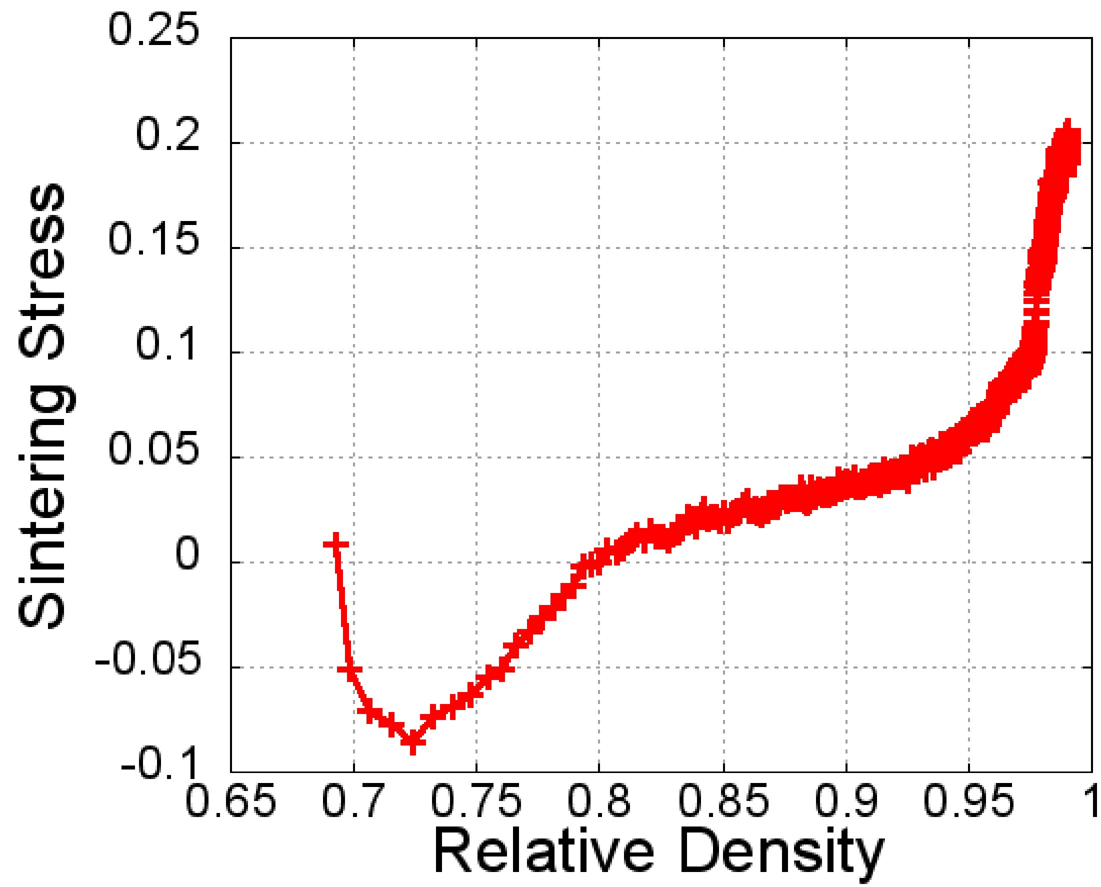
Faces = 0 **Edges**

  - **Digitized microstructure**



Sintering Stress
Simulation of Cu-Particle Compact Sintering

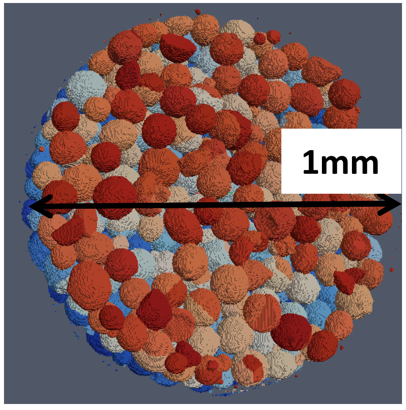




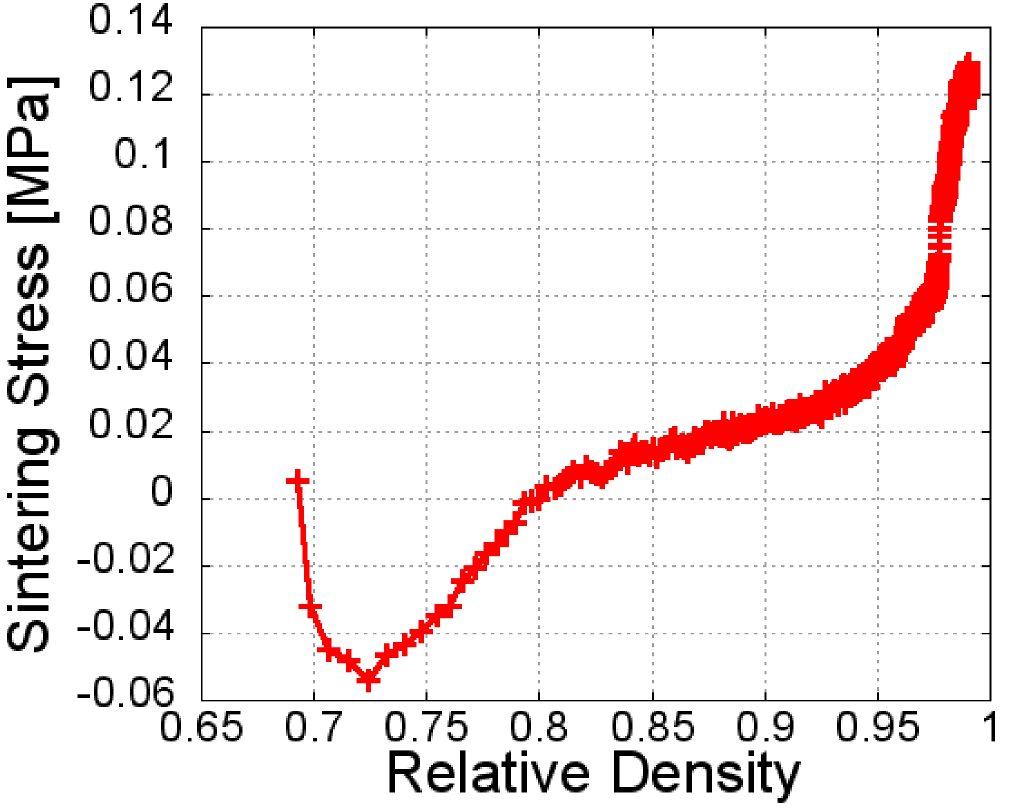
Sintering Stress
Simulation of Cu-Particle Compact Sintering

Obtaining P_L with units for real systems

•units of spatial dimension



•Assume γ_s = 1 J/m^2 at 1050 ^{o}C



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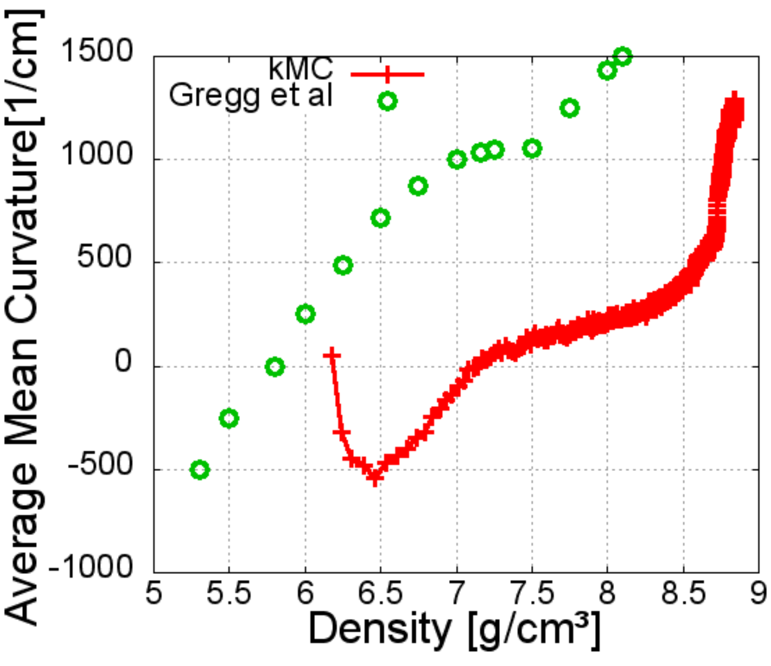
Sintering Stress, Comparison to Experiment
Simulation of Cu-Particle Compact Sintering



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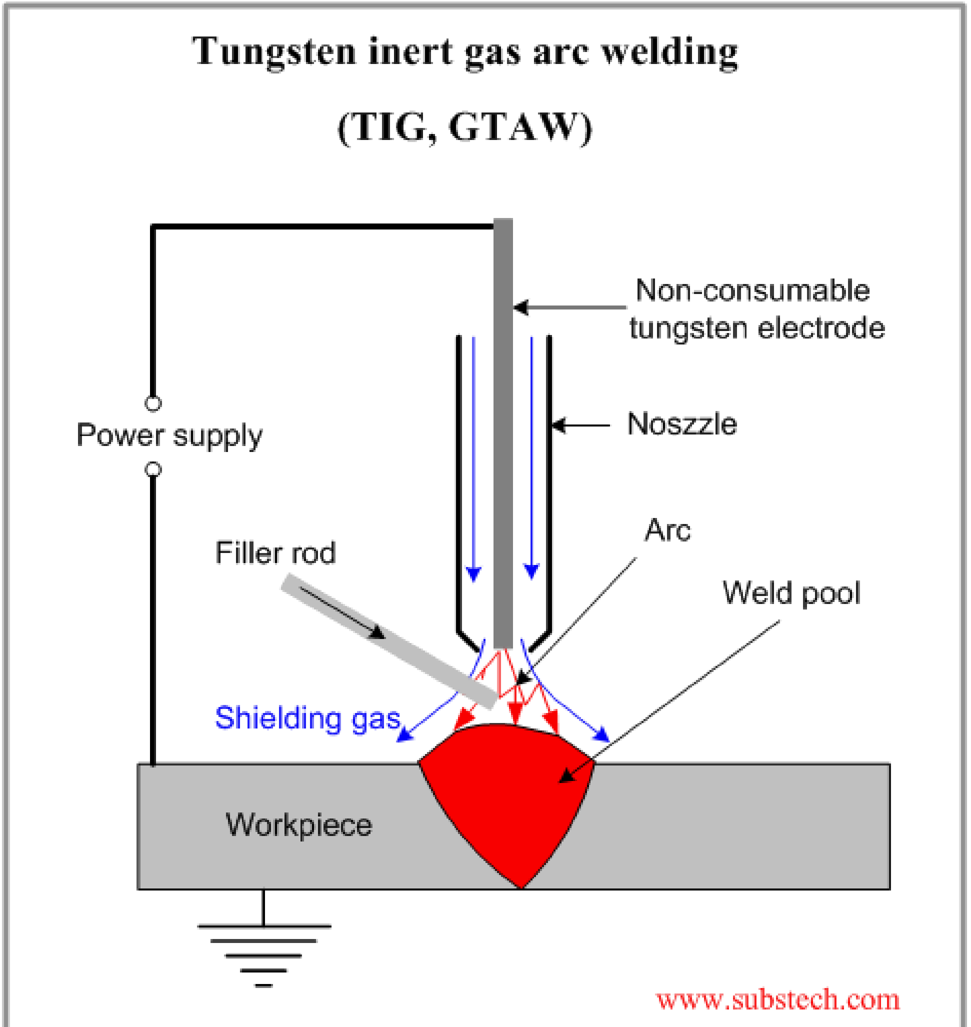
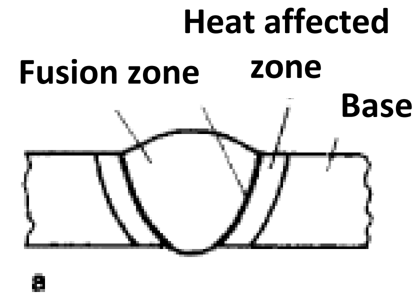
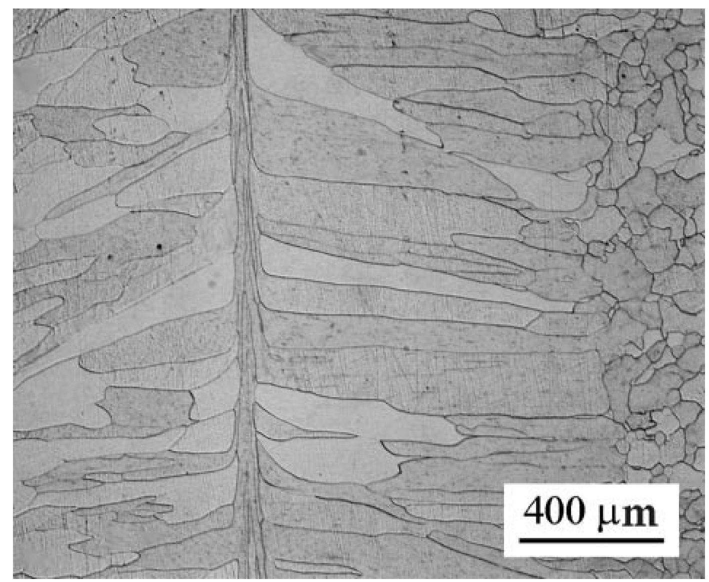
Welding and Resulting Microstructures





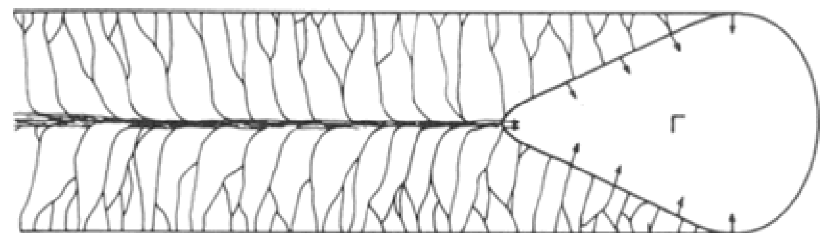


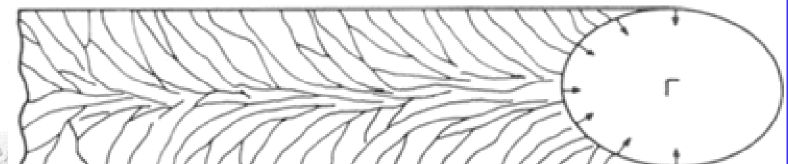
McKamey et al., S&T of Welding & Joining, 2000

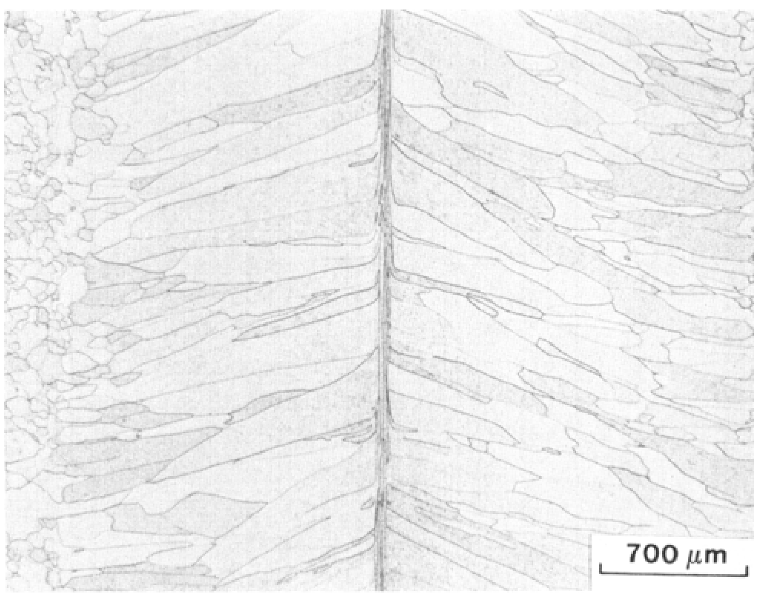
Welding and Resulting Microstructures

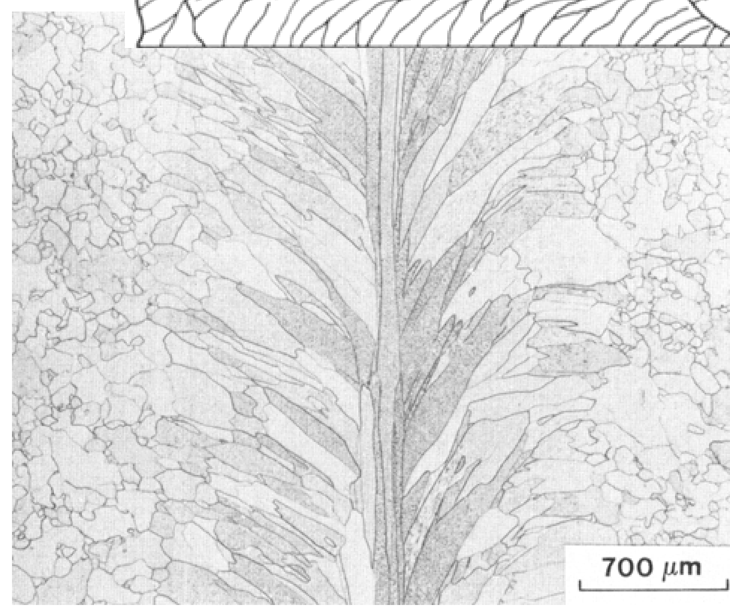
Welding of Iridium, DPO-26
Microstructure is function of

- bead width: 3.7, 2.5 mm
- Heat input
- Weld speed









Liu & David, Met. Trans A, 1982

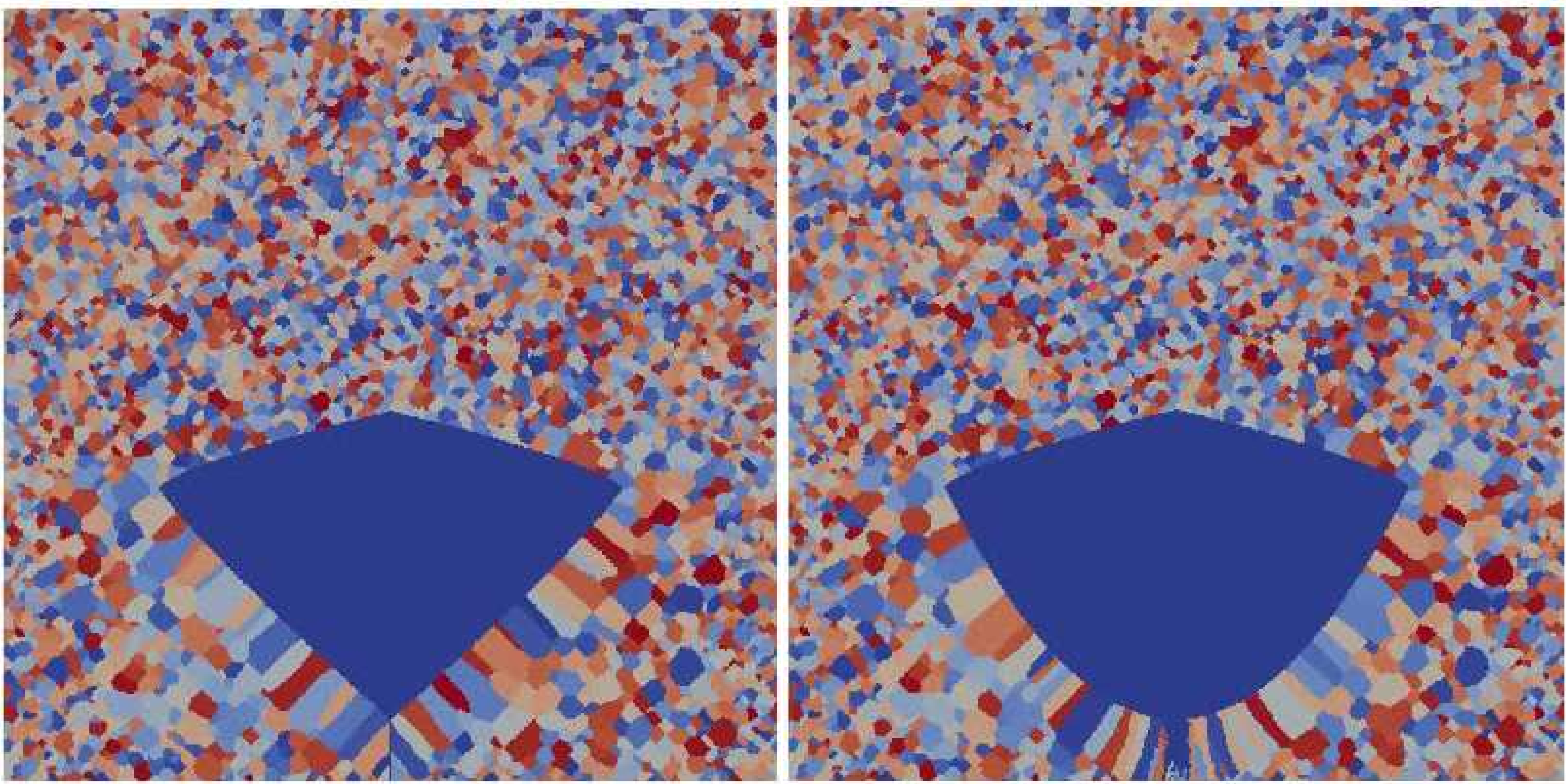
Preliminary μGEM Results, Weld Microstructures

Base metal

HAZ

FZ

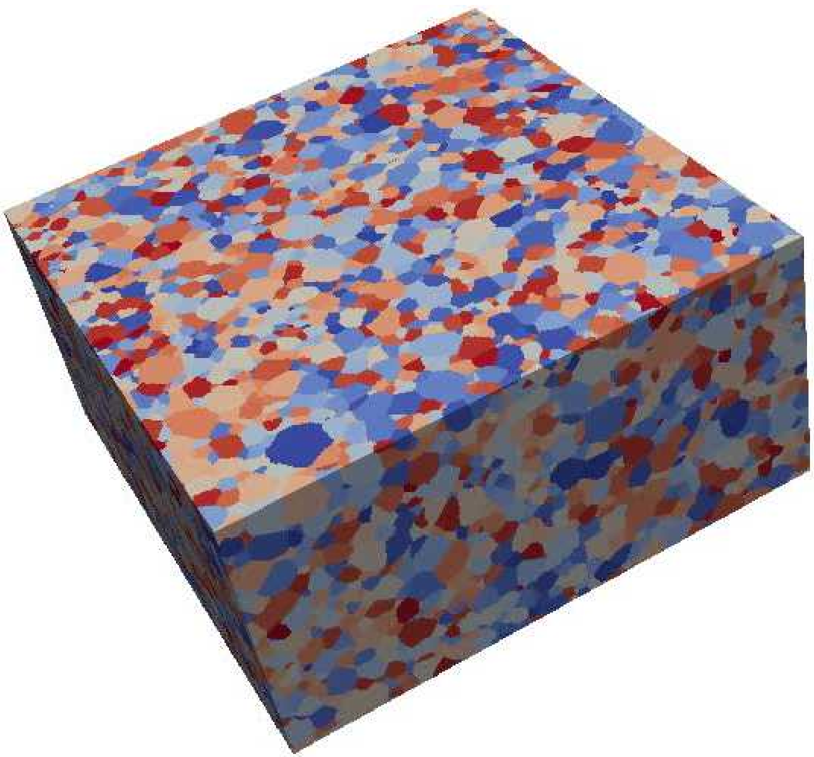
Grains grow epitaxially, perpendicular to iso-temperature lines

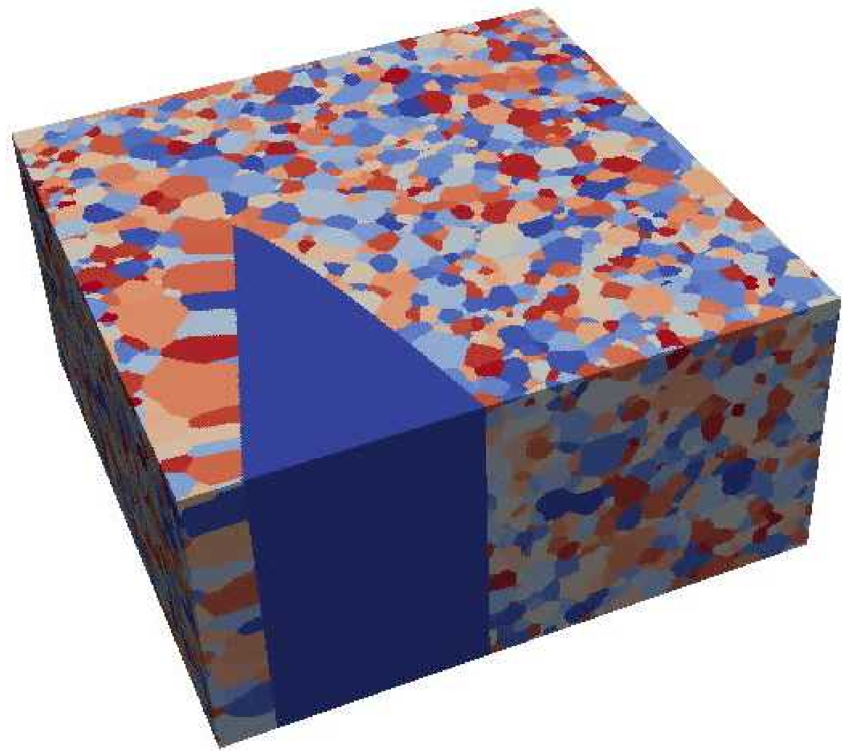


Preliminary μGEM Results, Weld Microstructures

3D simulations: complete grain size and shape evolution

- **Welding rate, 1 and 5x.**

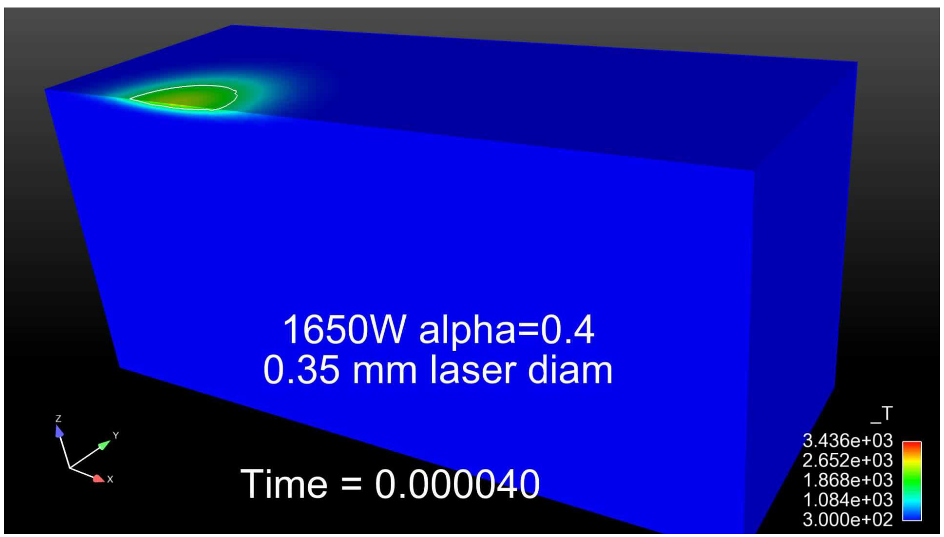




3D Traveling Laser Seam Welds

ARIA/CDFEM Simulation
Weld Speed: 20 cm/sec
1650 W 40% absorption
0.35 mm beam radius

Molten Weld Pool Dynamics
Laser Speed = 20 cm/sec



MJ Martinez, SNL

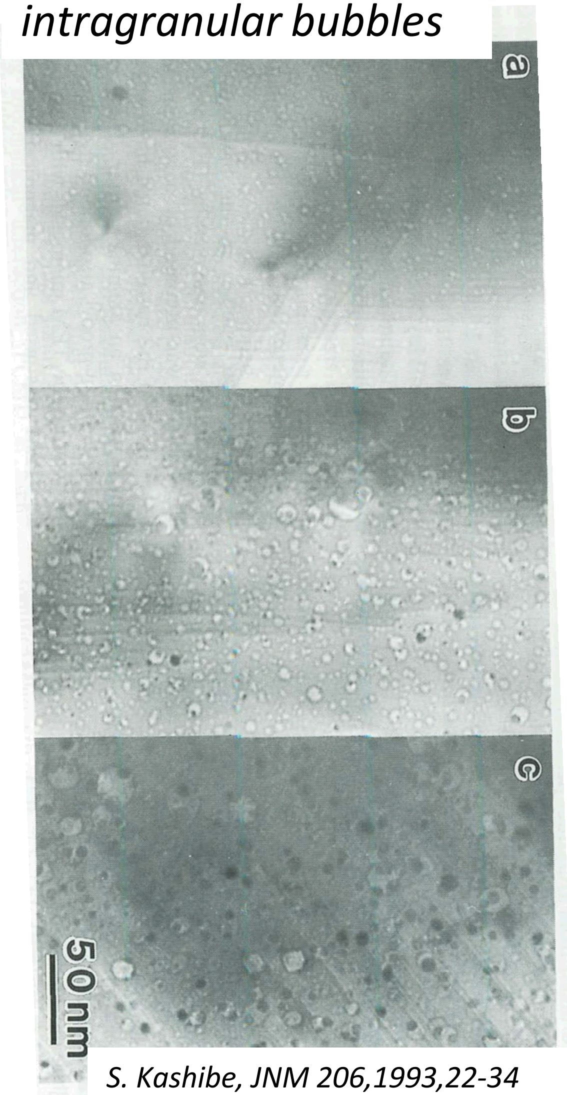
Model Features:

- Free-surface motion (ALE & LS)
- Free-surface BC
  - Surface tension & curvature
  - "Marangoni" stresses, $\sigma(T, C_i)$
- Melting/solidification
- Laser ablation on free-surface – nonequilibrium thermodyn. subgrid model
  - Ablation/evaporation energy loss
  - Recoil pressure
- Latent heat
- General fluid properties - k(T), Cp(T), μ(T)

M. Martinez, 2014

Gaseous Fission Products Accumulation & Release

intragranular bubbles



S. Kashibe, JNM 206,1993,22-34

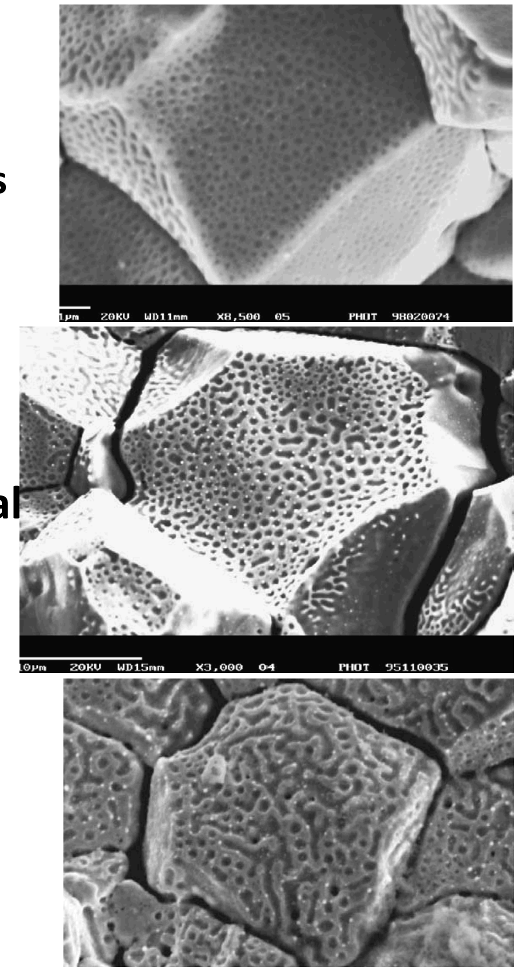
Fission gas is stored as

- **Dissolved gas in the fuel lattice**
- **Nano-sized intragranular bubbles**
- **Micron-sized intergranular bubbles**

Fission gas released by

- **Diffusion to surface**
- **Recoil & knock-out**
- **By percolation of the intergranular bubbles to external surface or cracks, Occurs in bursts.**

intergranular bubbles



R.J. White, JNM 325,2004,61–77

The model simulates fission gas generation and transport

Reality

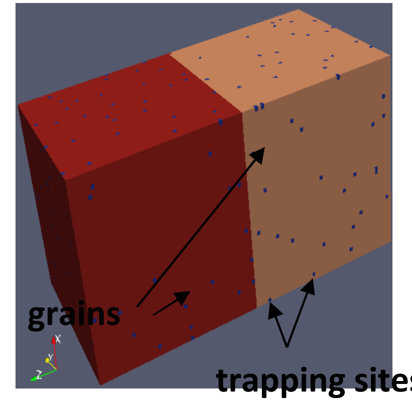
- **Fission gases are generated in proportion to fission events**
- **They diffuse in the fuel lattice until precipitating in bubbles**
- **Two sets of bubbles: inter- and intra-granular bubbles**
- **They precipitate on trapping sites (defects due to irradiation) to form nano-sized intragranular bubbles**
- **They precipitate on grain boundaries to form grain boundary bubbles**
- **Fission fragments and neutron irradiation re-dissolve the gases from the bubbles.**

Model

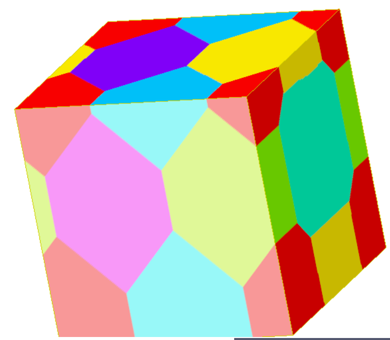
- **Gas atoms are generated in proportion to fission events**
- **They diffuse by random walk with D(T, damage, C, …)**
- **Two sets of bubbles: inter- and intra-granular bubbles**
- **They precipitate on trapping sites (stationary nucleation points) and diffuse along the surface**
- **They precipitate on grain boundaries and diffuse and coalesce on the gb to form bubbles**
- **Fission fragments and neutron irradiation re-dissolve the gases from the bubbles.**

Model developed in stages by applying to increasingly more complex geometries

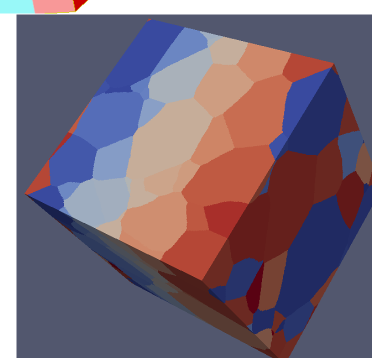
1. **Two grain geometry with only grain boundary faces**
   - Used to develop simulation parameters
   - Study percolation on planar grain boundary with realistic bubble topology



2. **Close packing of tetrakaidecahedral grains**
   - Regular grain faces
   - Grain edges and corners

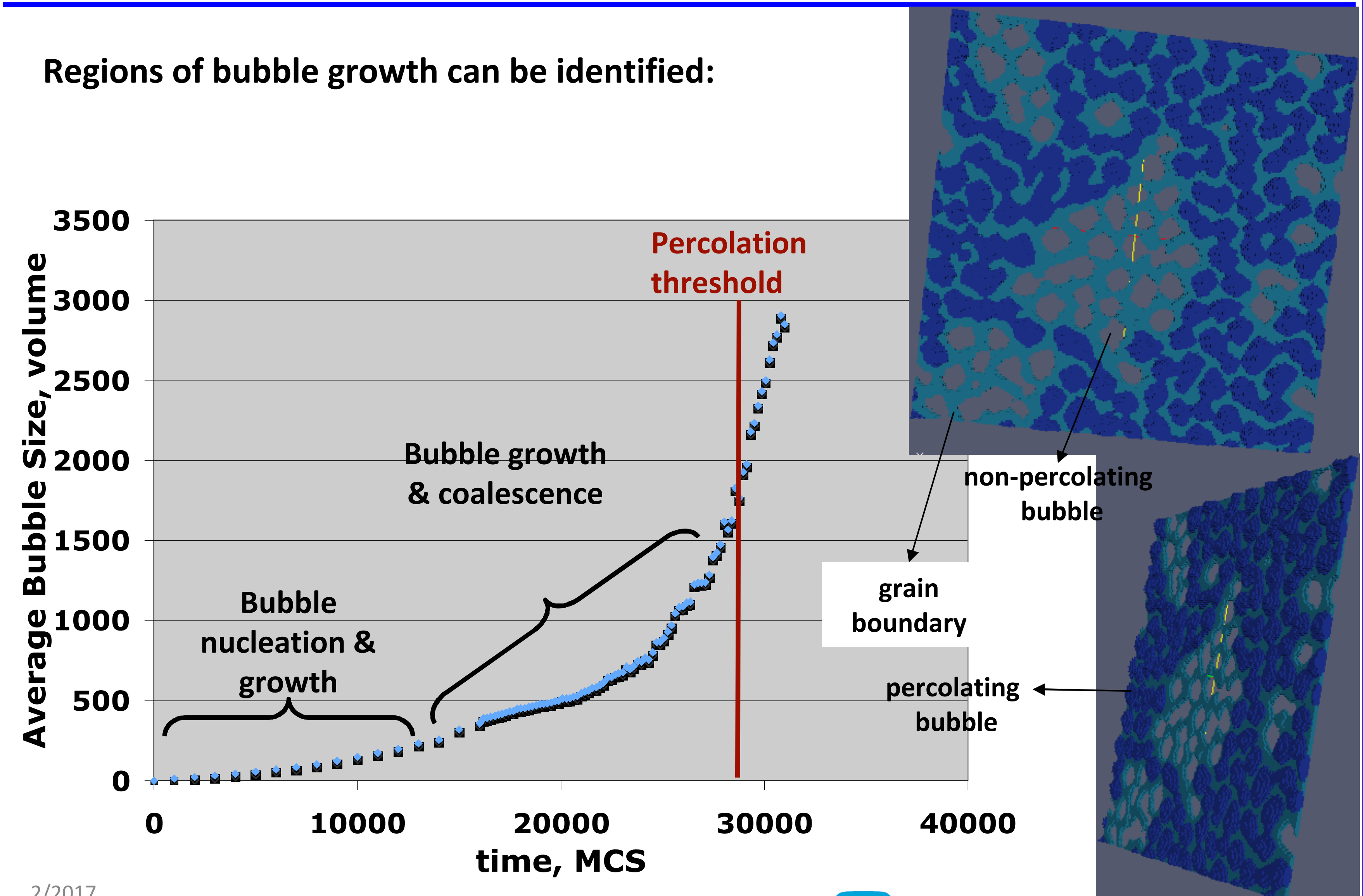


3. **Topologically correct polycrystalline geometry**
   - Introduce all the complexity of polycrystalline grains
   - Faces of different size, shapes curvatures,
   - edges of different lengths and curvatures
   - corners at varying distances

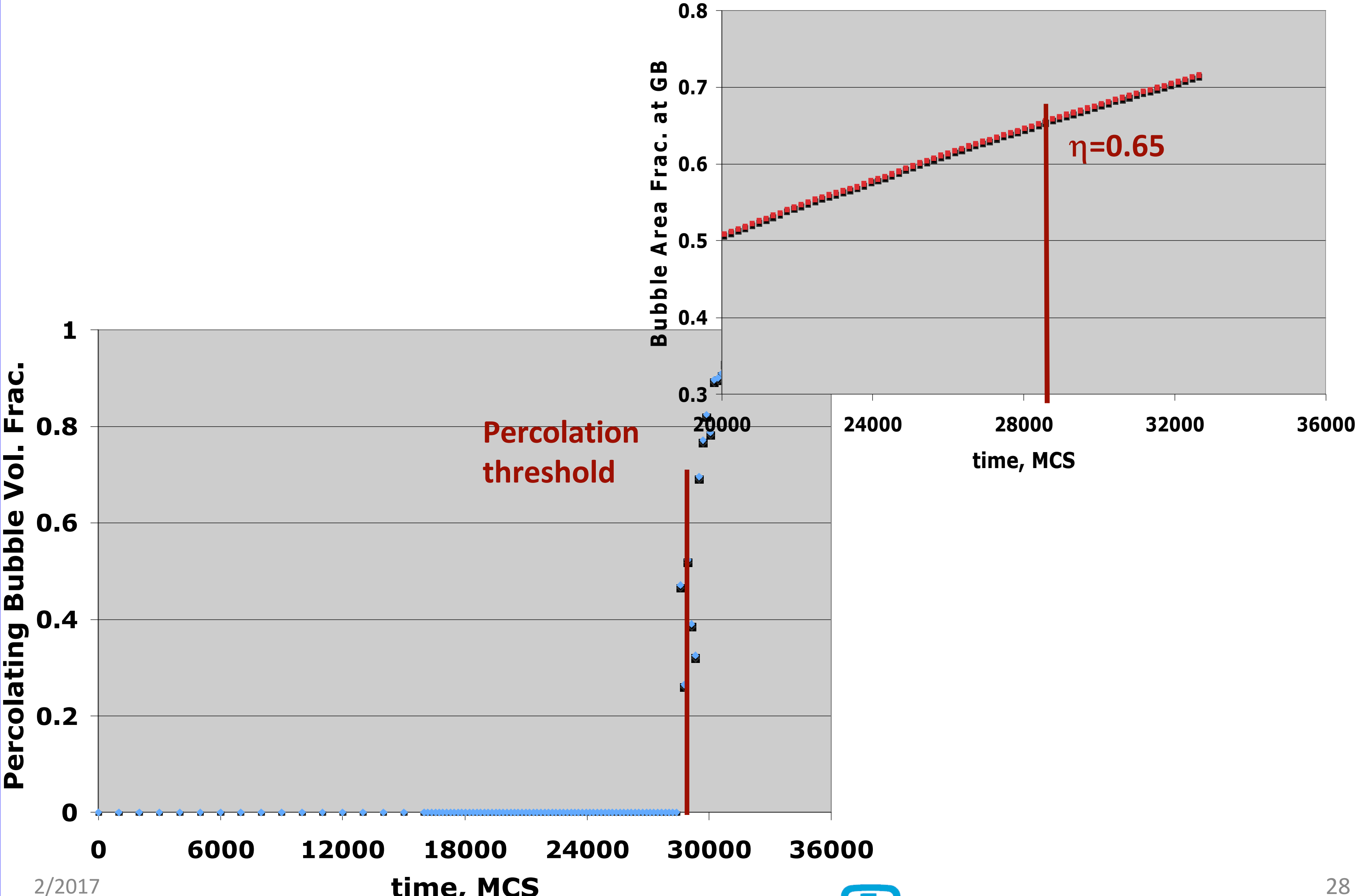


Bubble growth at semi-infinity grain boundary

Regions of bubble growth can be identified:



Bubble growth at grain boundary



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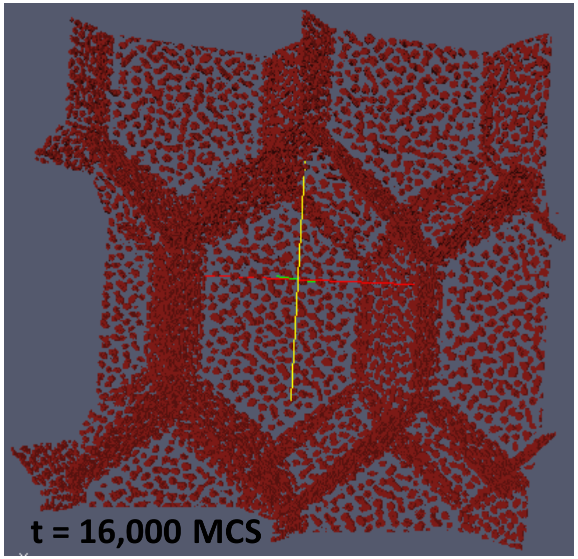
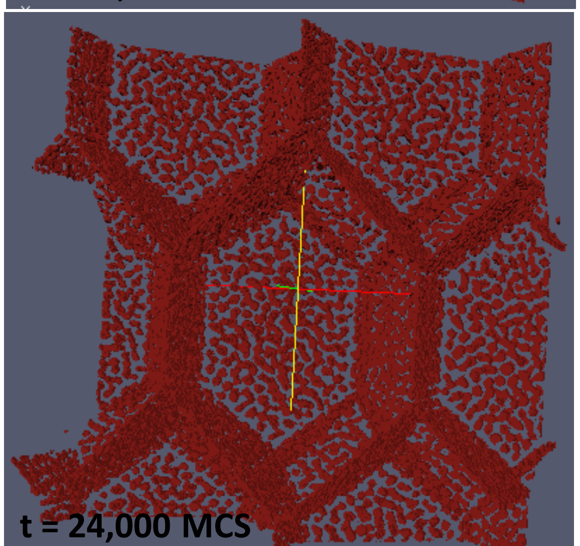
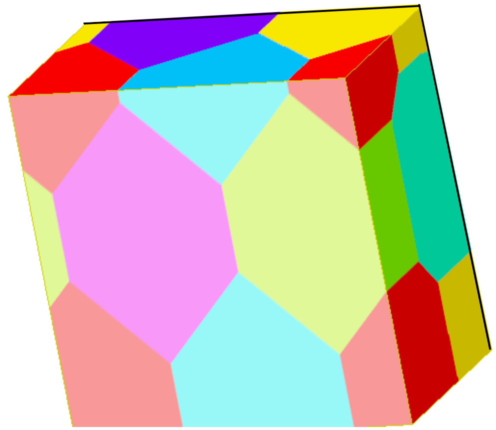
Fission Gas Bubble Behavior in CP TK Grains

Bubbles at GBs

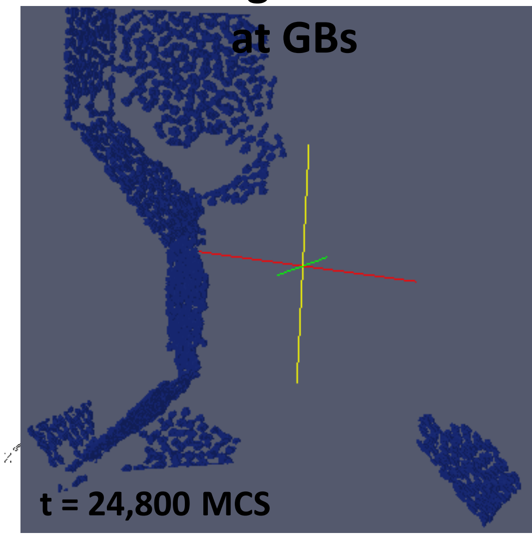


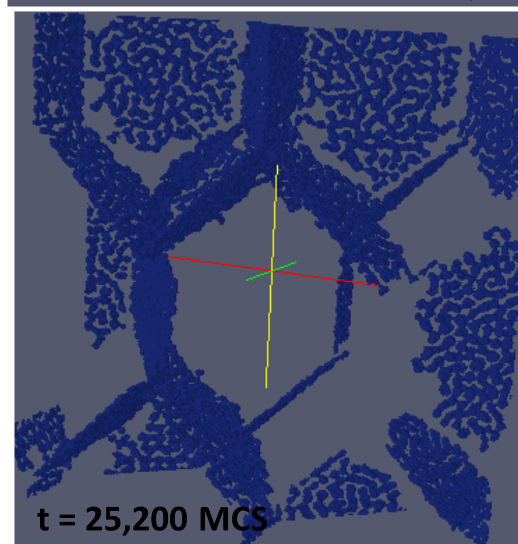


Half the cube is imaged



Percolating bubbles at GBs

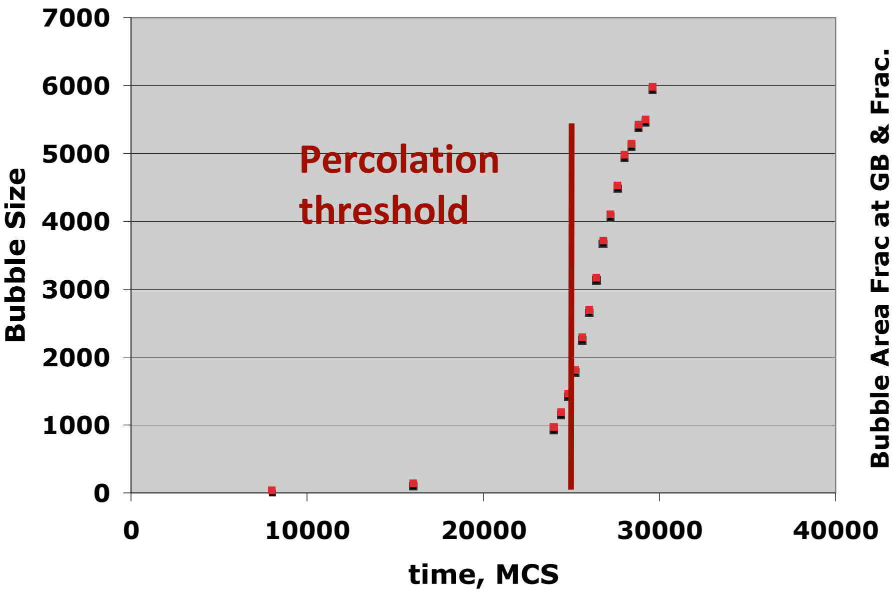


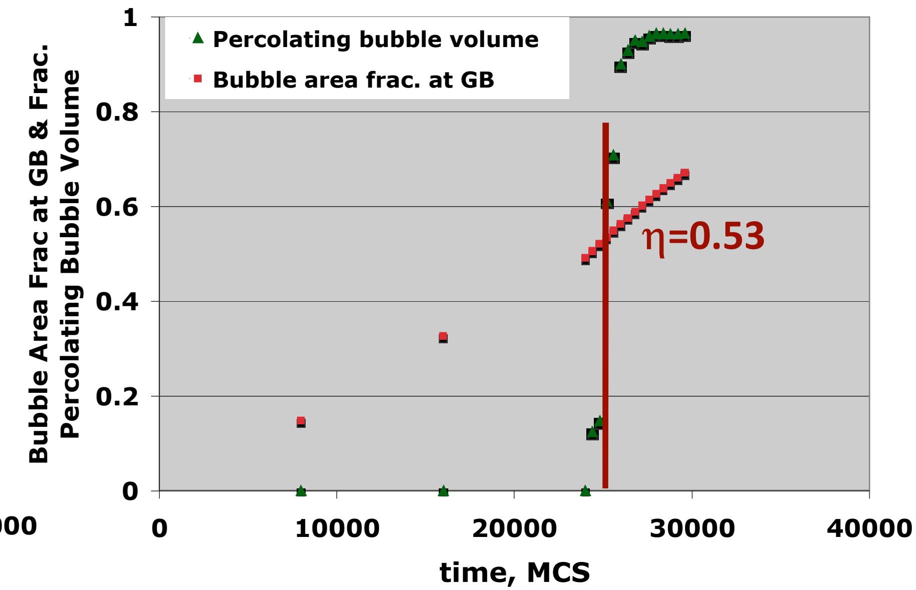


The same 3 regions
- **nucleation & growth**
- **growth & coalescence**
- **percolation**

are observed in poly-crystalline fuel grain boundaries.

Fission Gas Bubble Behavior in Tetrakaidecahedral Grains





The polycrystalline fuel grain boundary bubbles behaved similarly to the bi-grains with infinite grain boundaries.

- **3 regions of intergranular bubble growth**
- **Percolated at *lower gb coverage η=0.53***
- **Grain edges lower percolation threshold**

Fission Gas Bubble Behavior in Polycrystalline Grained-Fuels

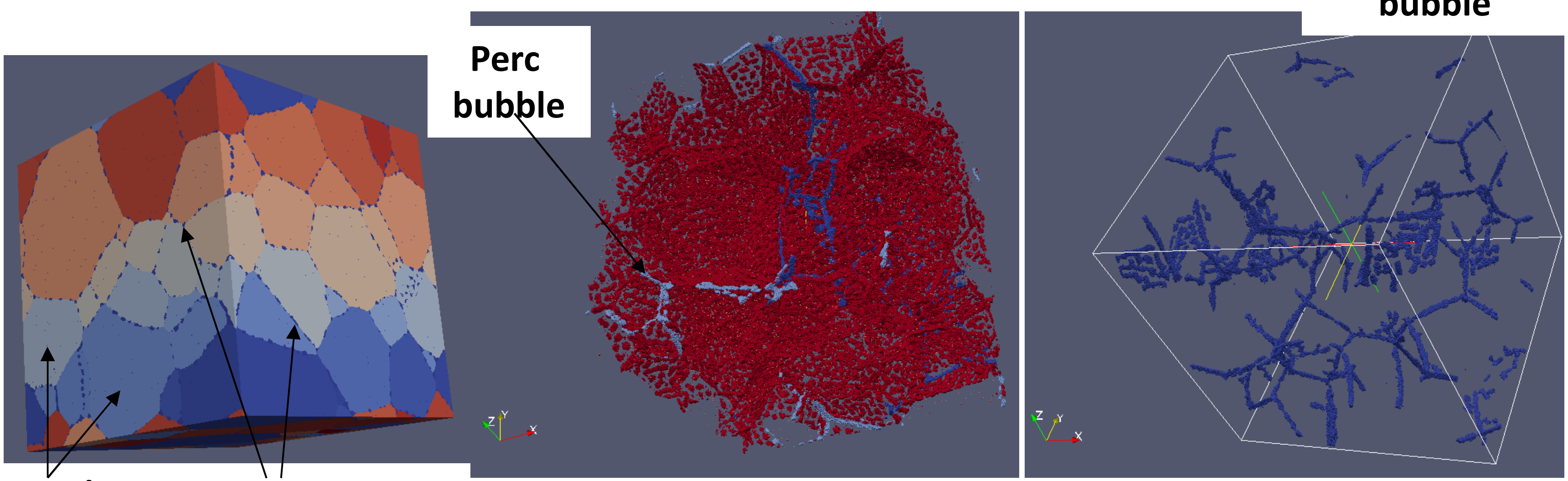
Bubble growth and percolation in a polycrystalline fuel

- with distribution in face and edge size and shape.

- Bubble shape at edge is ellipsoidal
- Bubble shape at corners is tetrahedral
- Linkage among edge & corner bubbles occurs before that on grain boundary faces

All bubbles

Percolating bubble

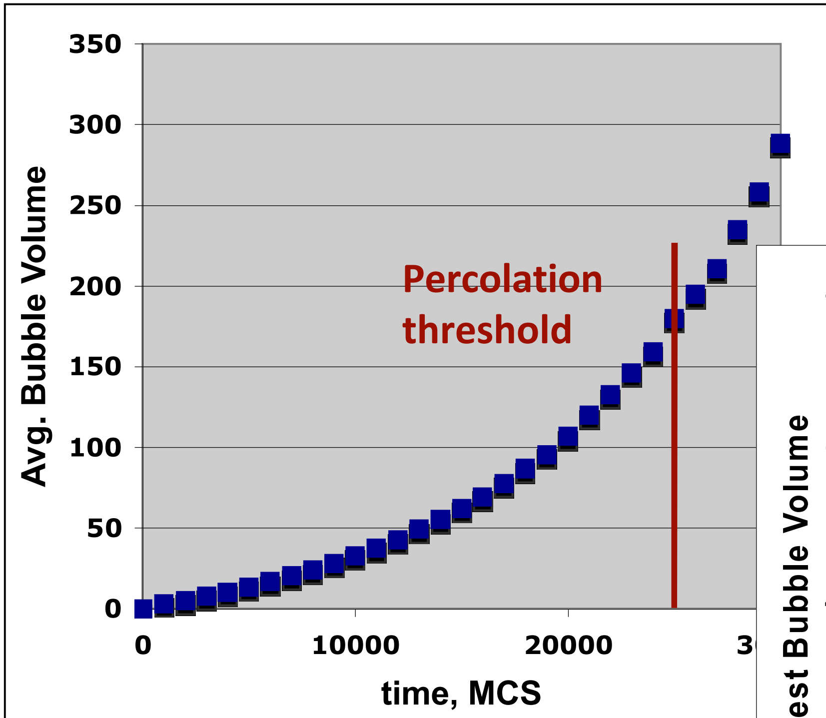


grains bubbles

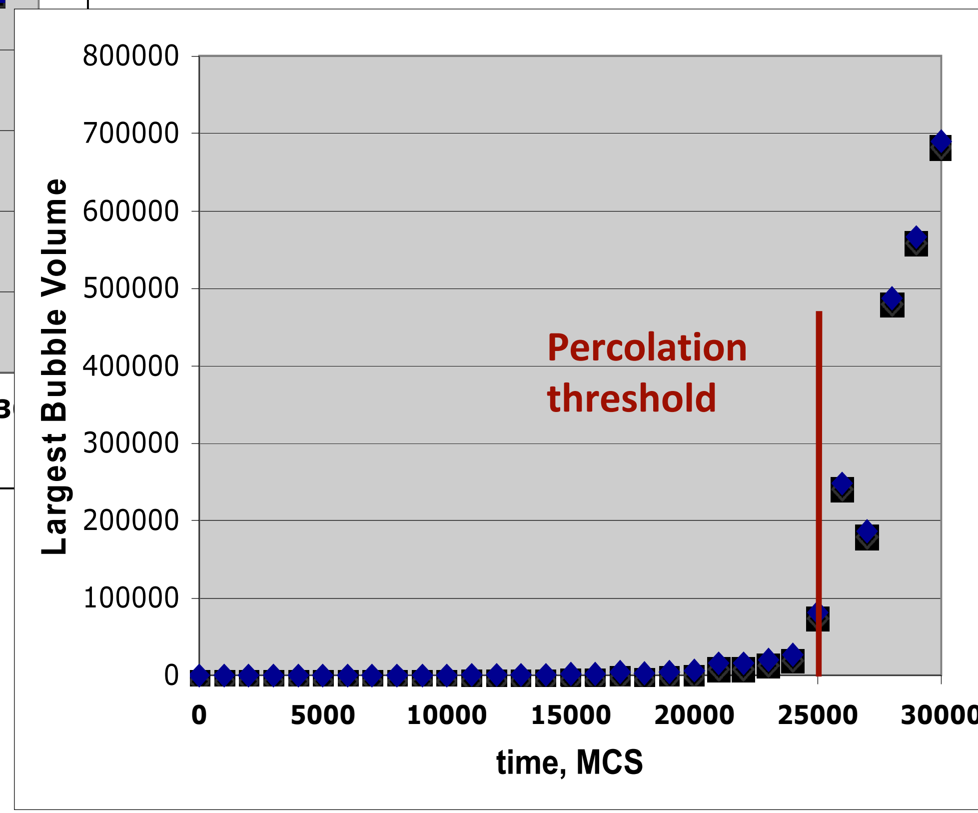
t = 24,000 MCS

t = 25,000 MCS

Fission Gas Bubble Behavior in Polycrystalline Grained Fuels



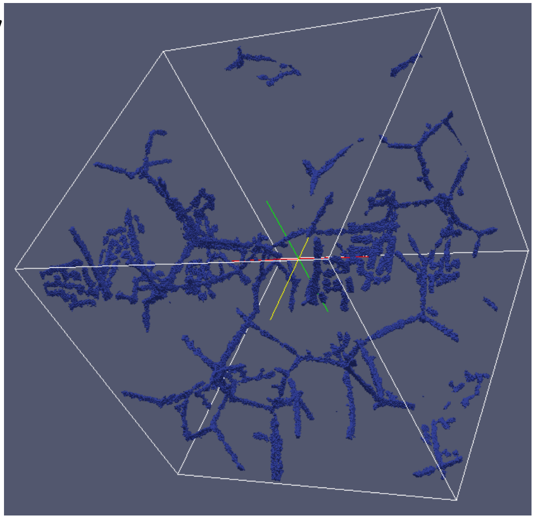
Grain boundary coverage at percolation is $\eta = 0.47$



Fraction of gas in gb bubbles released is $\psi = 0.057$

Fission gas in percolating bubble is released, sinters and bubble growth continues

- **The percolating bubble will release the gas within it as it is under pressure.**
- **Once the pressure is released, let us assume the region sinters back, so that no unpressurized porosity is left. The assumption here is that sintering is relatively fast compared to fission gas formation and diffusion.**
- **But, the non-percolating bubbles remain as they are pressurized.**
- **Continue simulation of gas formation and transport from this sintered geometry.**



The percolating bubble in the figure shown is allowed to sinter back.

Fission gas formation and transport in polycrystalline fuel with percolating bubble sintered

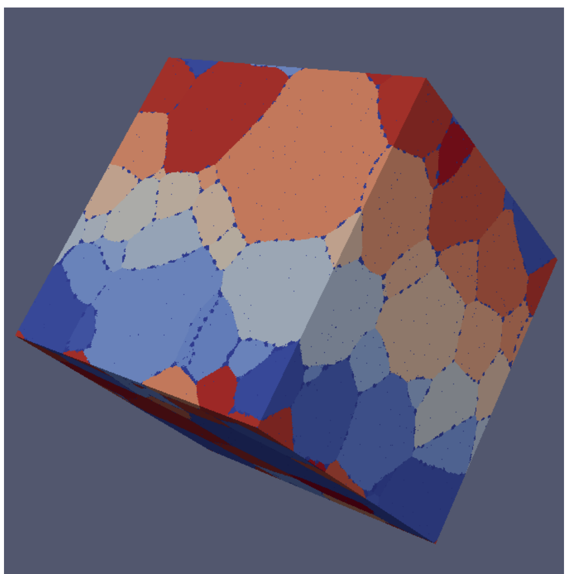
When percolation occurs, the polycrystalline has a large inventory of gas in gb bubbles.

The percolating bubble on the far right sinters and closes up, but all the other bubbles remain.

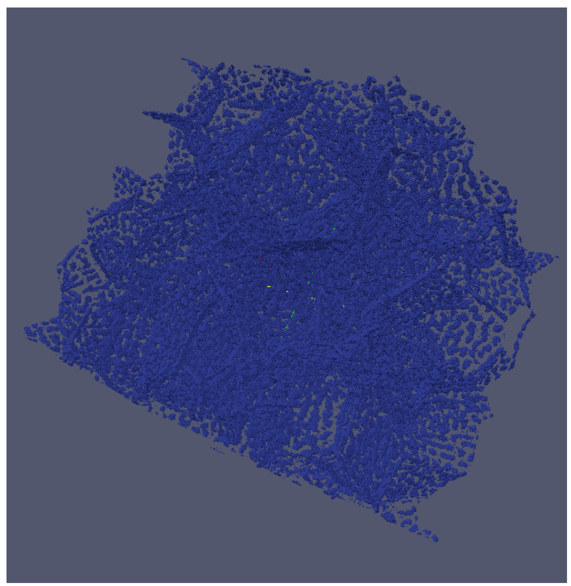
Gas generation and transport continue.

This bubble is sintered but the rest are left.

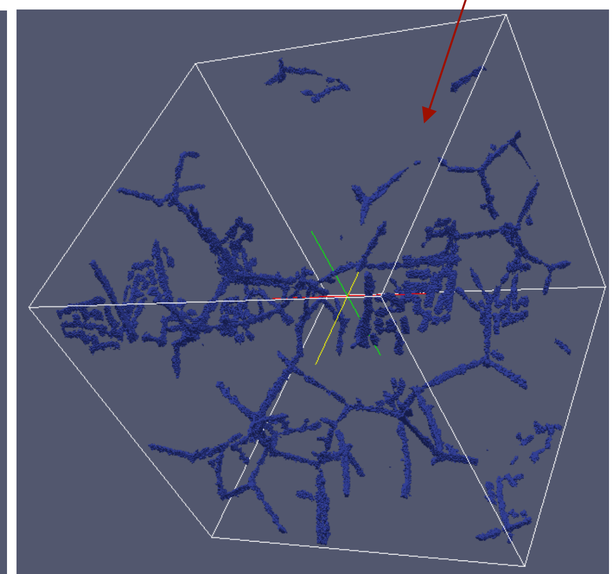
Gas bubbles in polycrystal



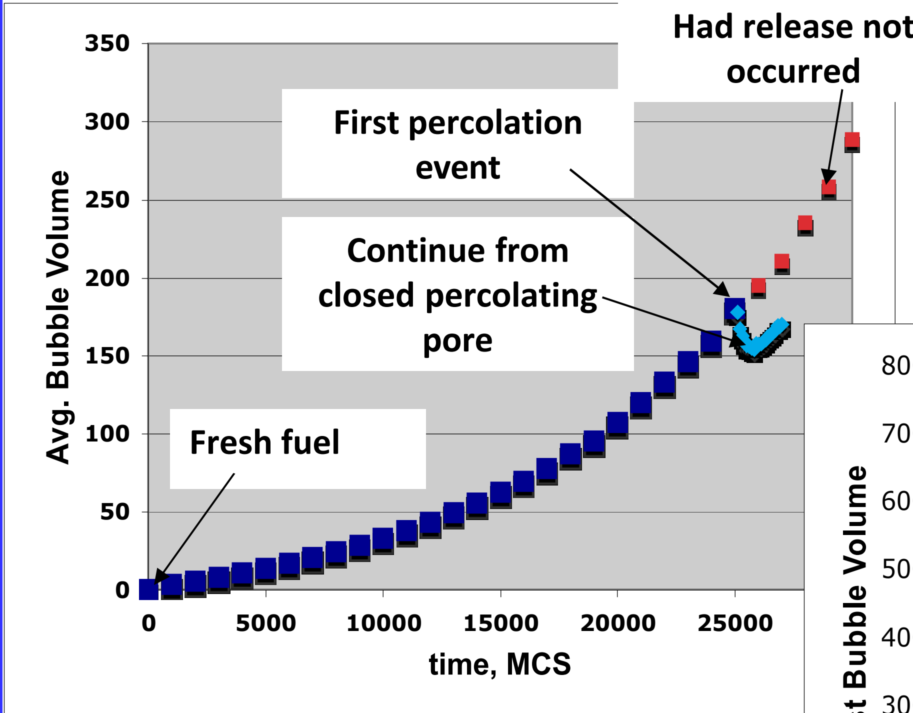
Gas bubbles only



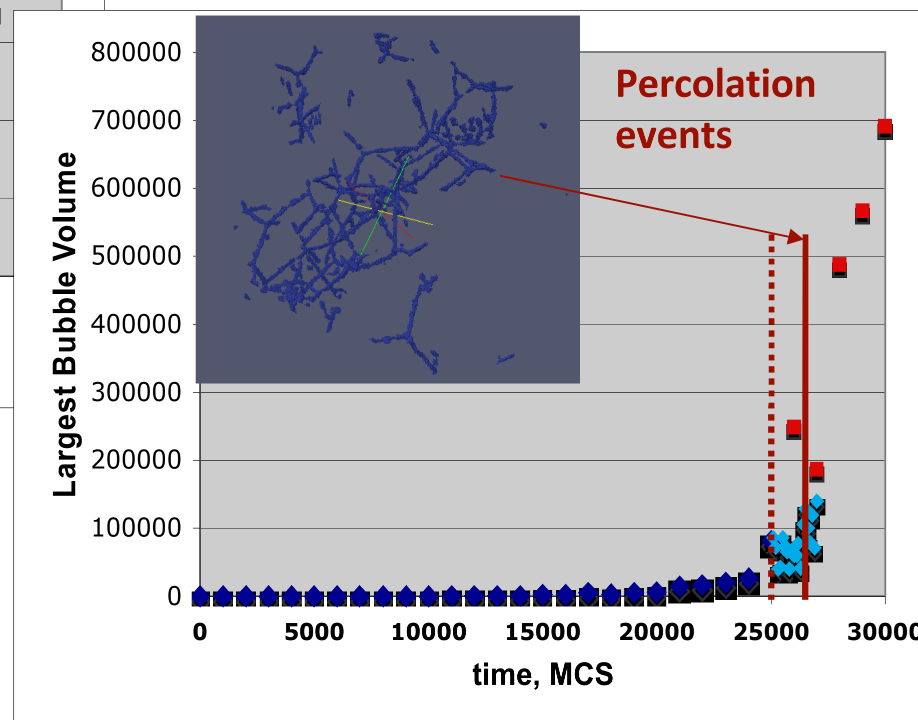
Percolating gas bubble only



Fission gas formation and transport in polycrystalline fuel with percolating bubble sintered



2nd percolation event follows quickly. Fraction of gas in gb bubbles released is ψ = 0.096 at gb coverage η = 0.44

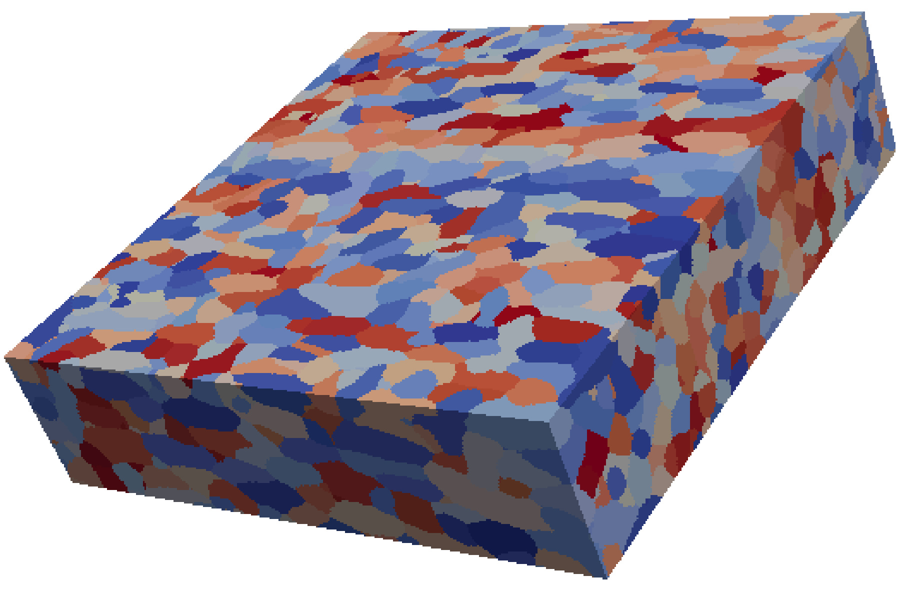


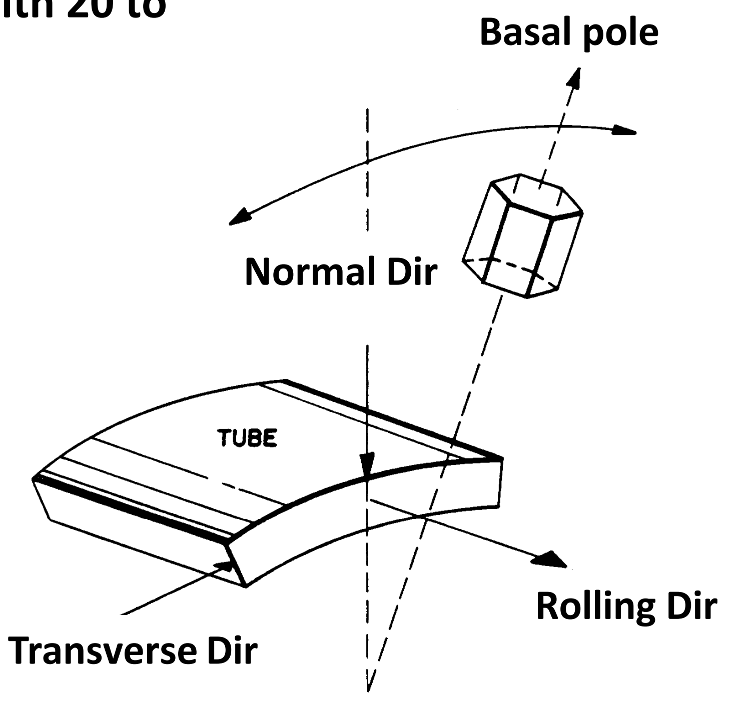
The average bubble size decreases slightly upon release as new bubble nucleate and grow in the sintered region.

Zircaloy-4 and other Zr-based claddings are known to have texture

Pilgering process imparts texture

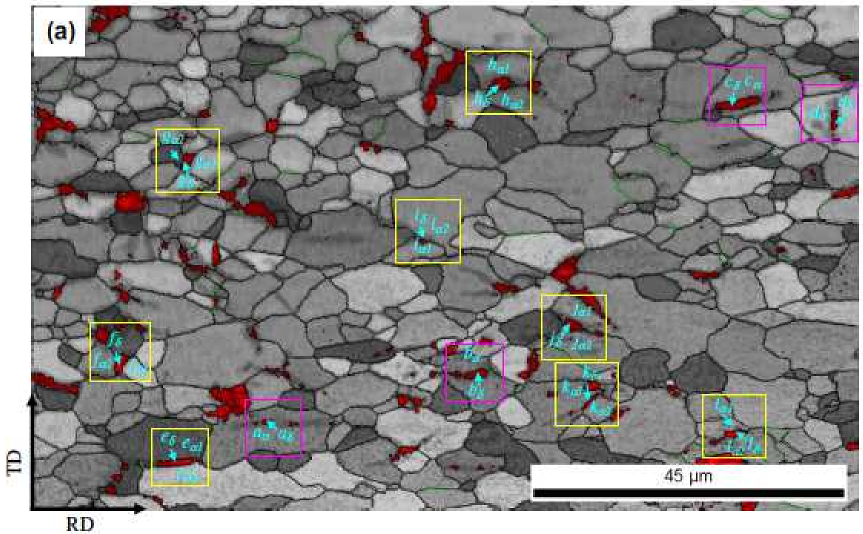
- Grain shape, elongated ~2x in rolling direction
- Crystallographic, basal plane parallel to ND with 20 to 40° rotation around TD

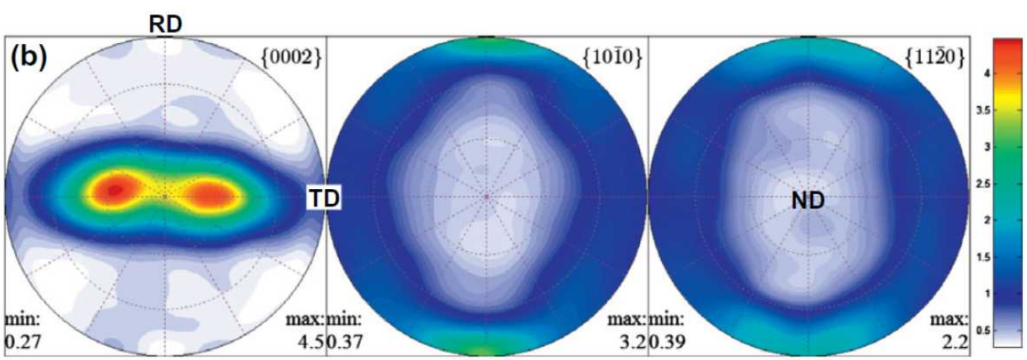


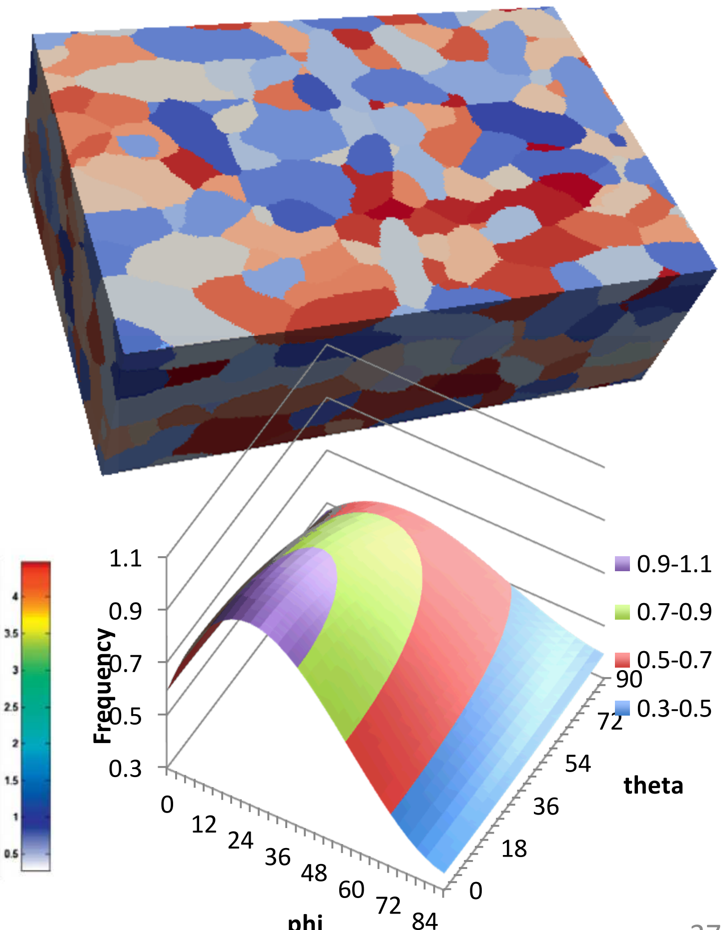


Generated a Digital Zr-4 Microstructure

from information available in the literature on grain and crystallographic texture

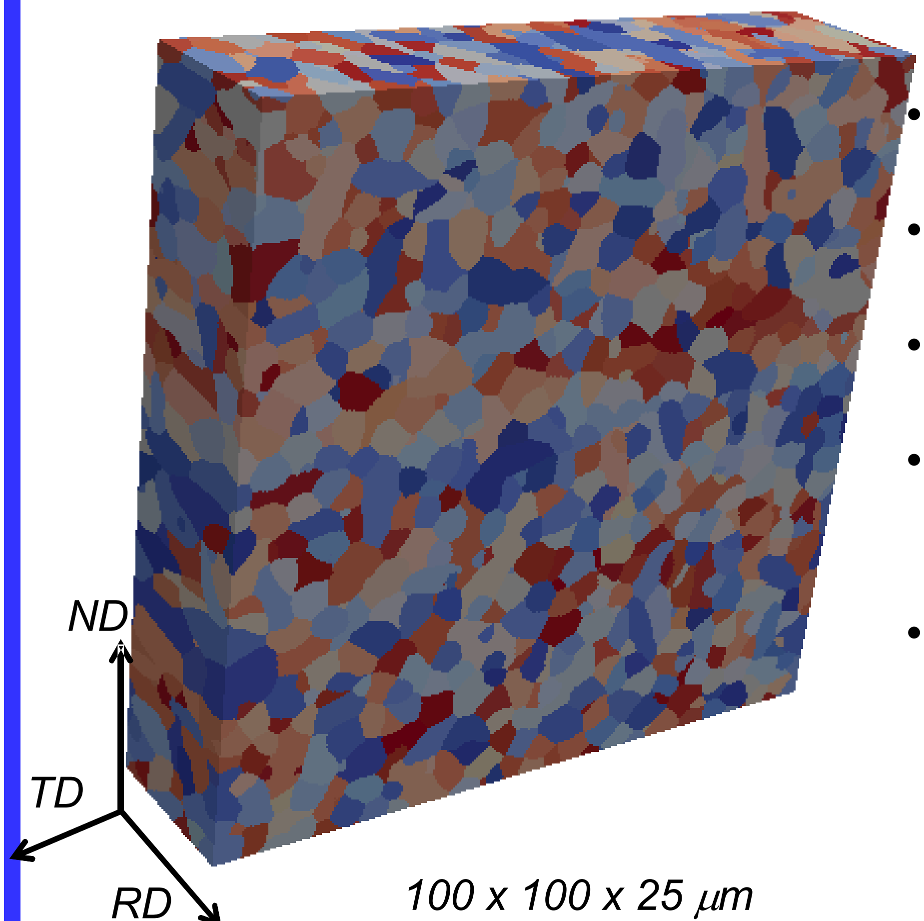






Sandia National Laboratories

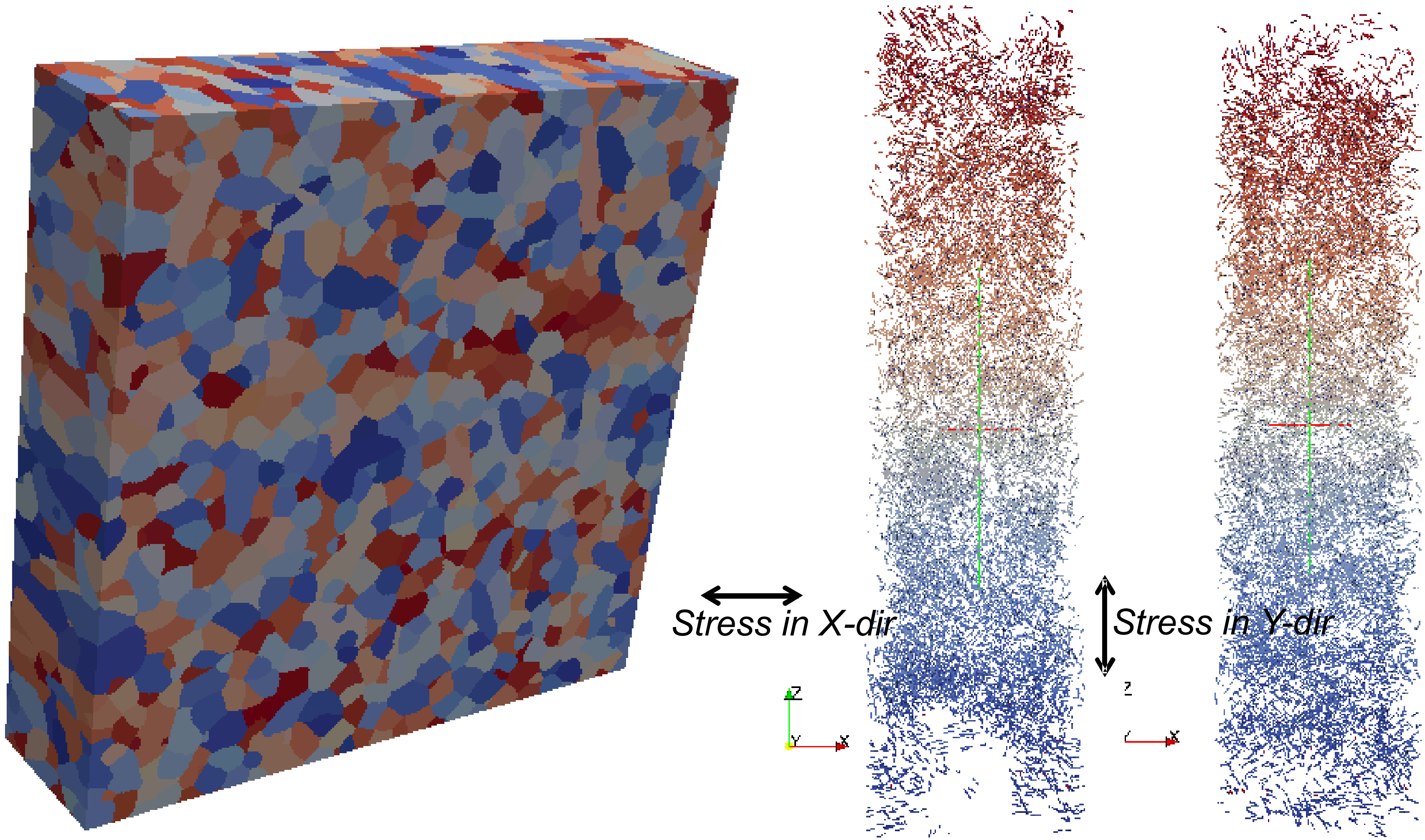
Simulation of d-$ZrH_{1.5}$ precipitation in rolled a-Zr with randomly oriented grains.



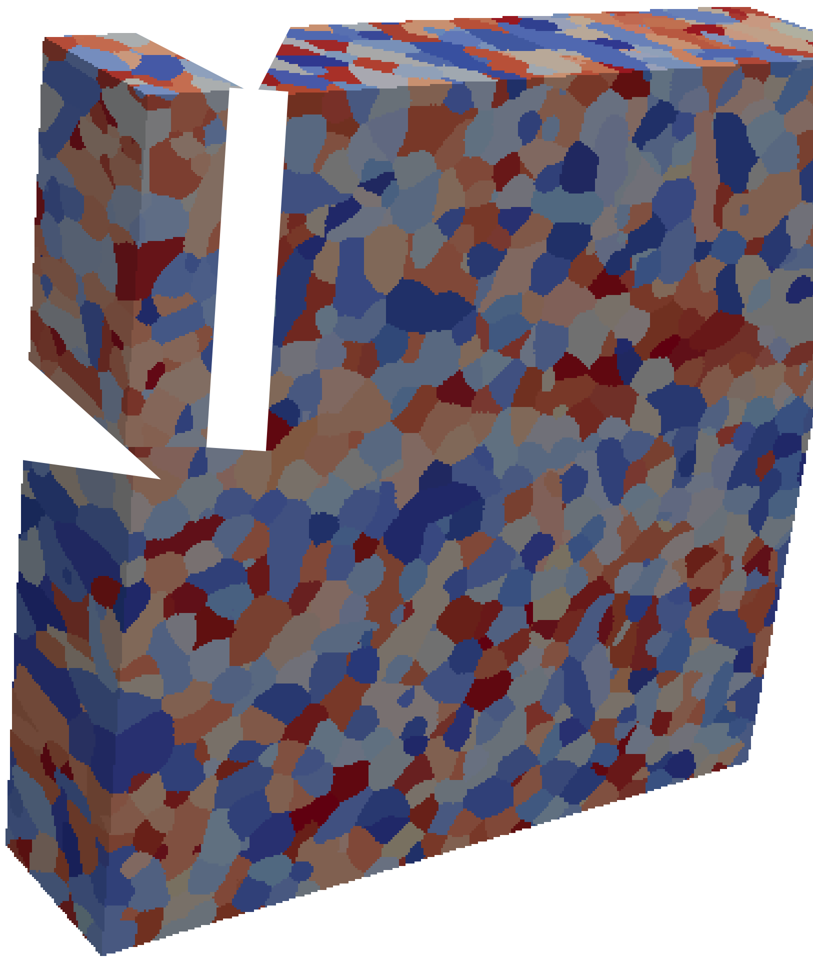
100 x 100 x 25 μm

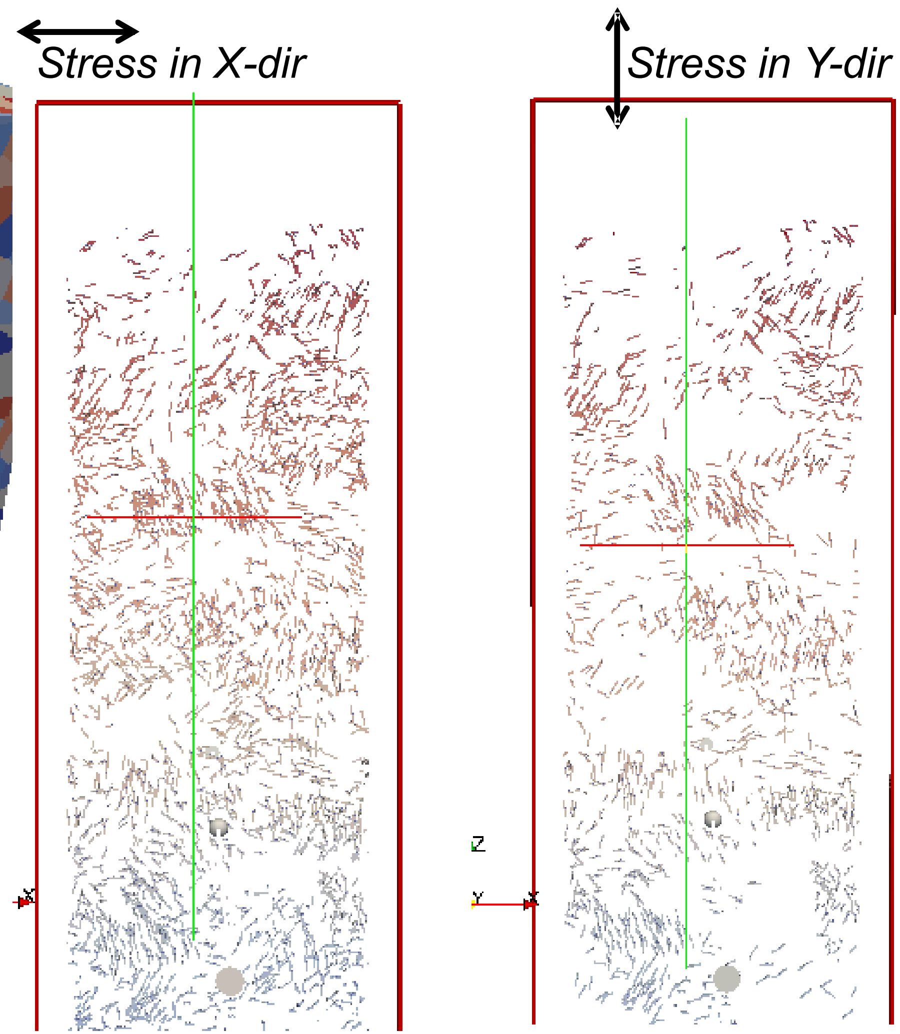
- Same microstructure as rolled Zircaloy-4 with elongation in rolled direction.
- Grain are randomly oriented with uniform distribution in all directions.
- Starting condition is supersaturated Zr with H, cooled to 350 ^{o}C.
- Precipitates form in basal plane with correct orientation δ(111) || α(0001), δ[1-10] || α(11-20)
- Nucleation at randomly selected sites with uniform distribution in each grain.

Simulation of d-$ZrH_{1.5}$ precipitation in rolled a-Zr with randomly oriented grains.

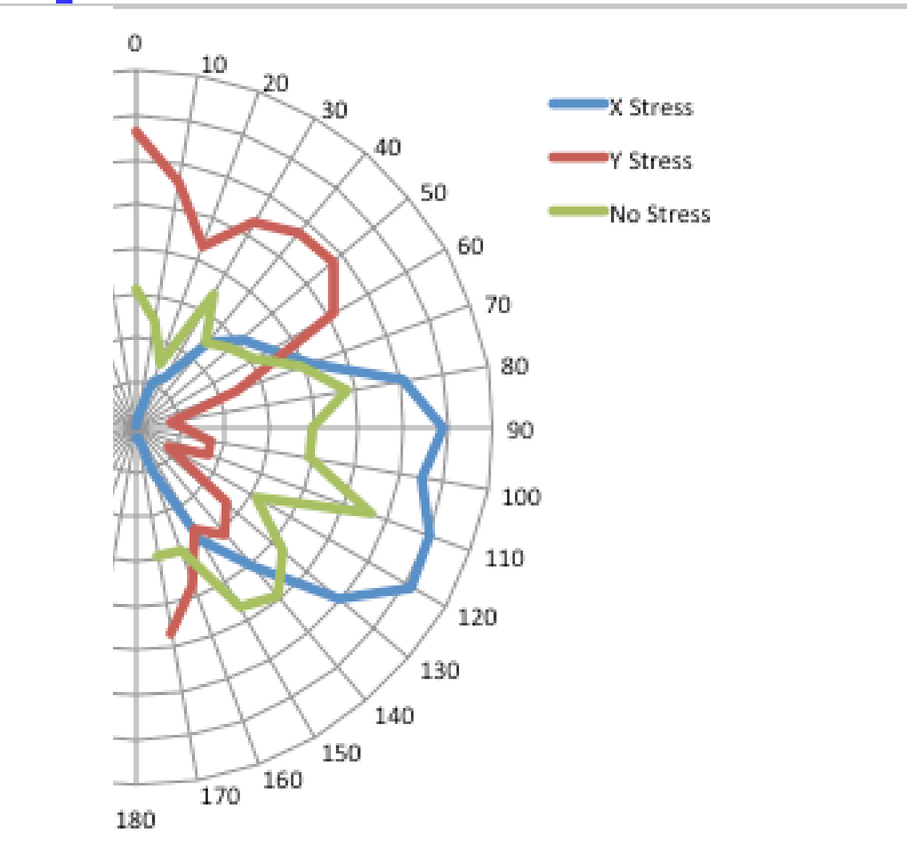


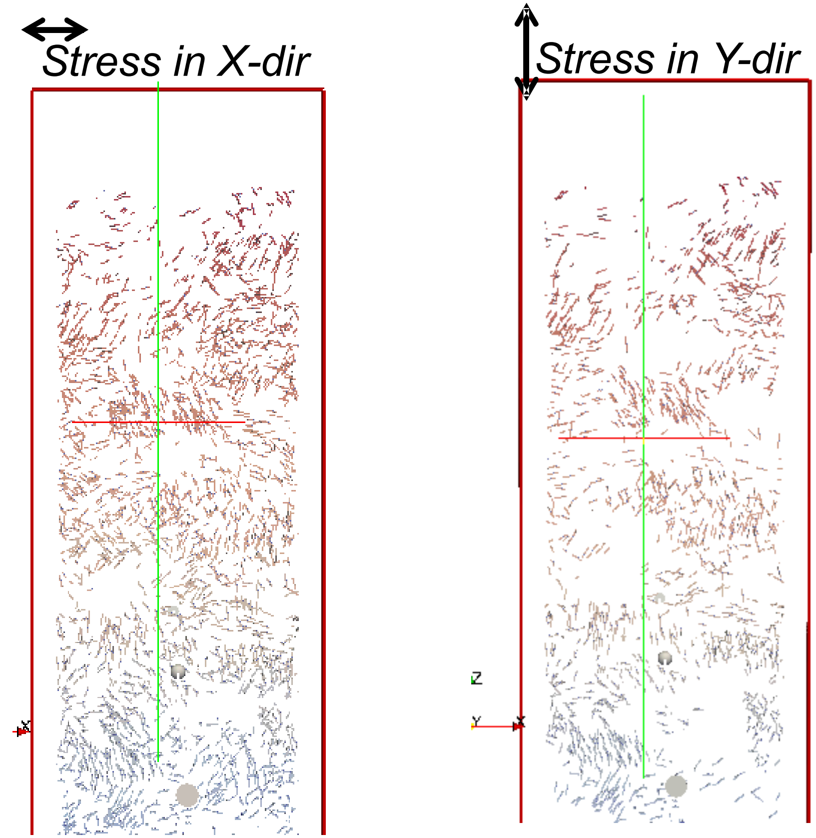
Simulation of d-$ZrH_{1.5}$ precipitation in rolled a-Zr with randomly oriented grains.





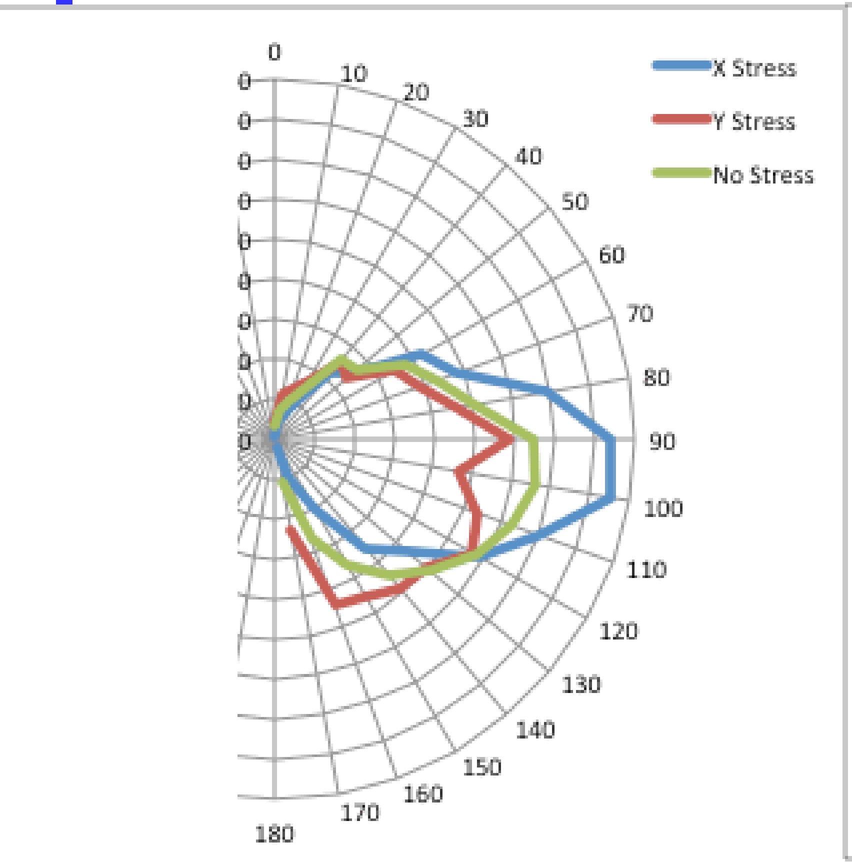
Simulation of d-$ZrH_{1.5}$ precipitation in rolled a-Zr with randomly oriented grains.



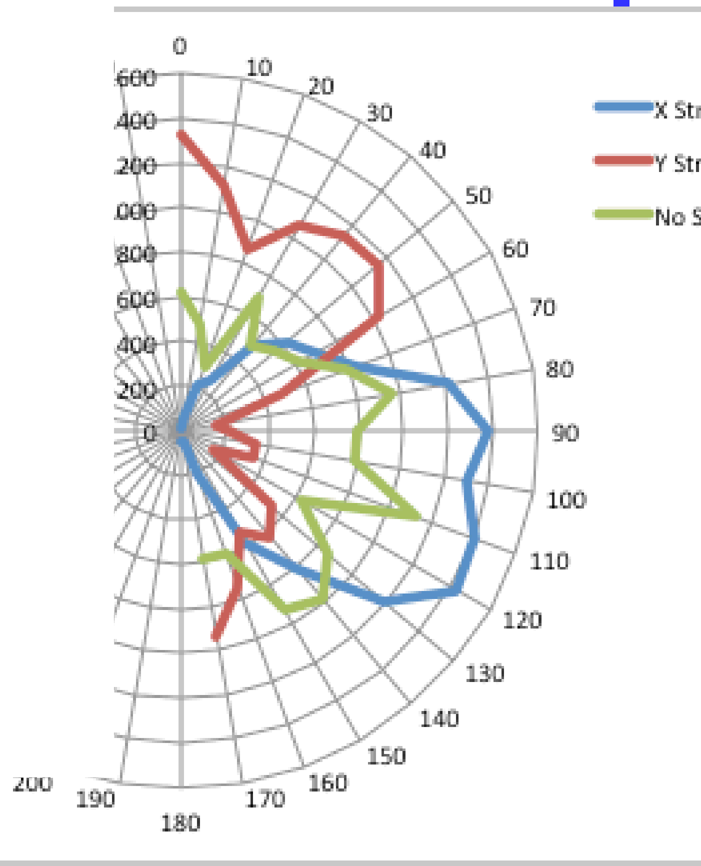


Reorientation occurs by precipitates in different grains that are favorably oriented precipitating to form "continuous" looking precipitates.

Simulation of d-$ZrH_{1.5}$ precipitation in rolled a-Zr with Zr-4 crystallographic texture.

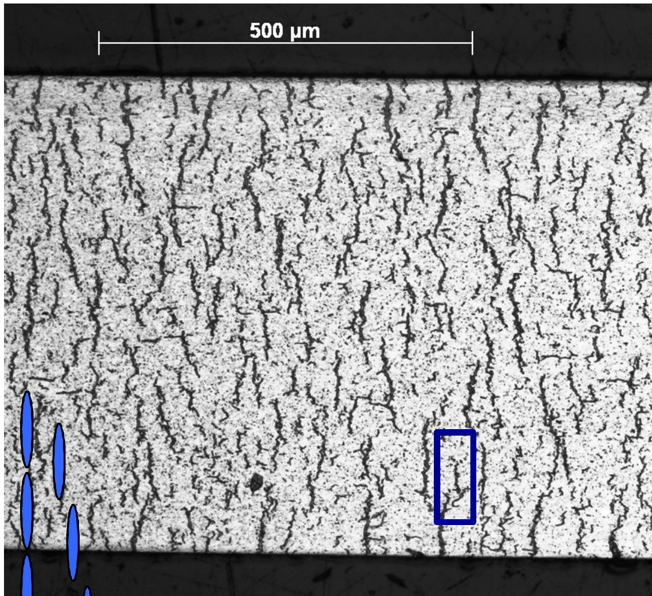


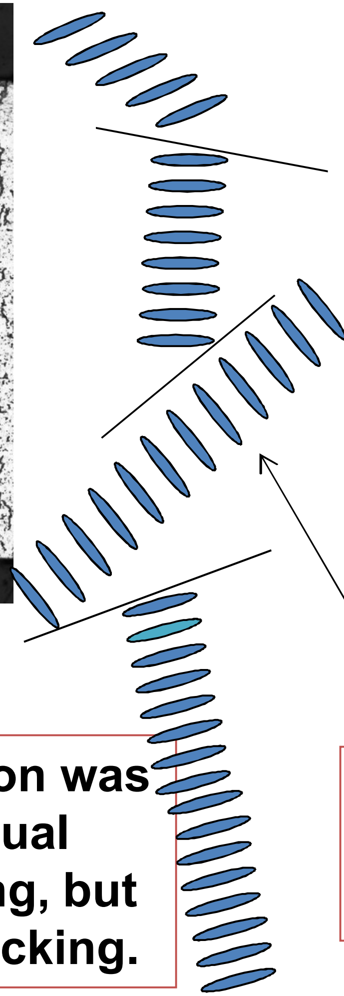
With crystallographic texture

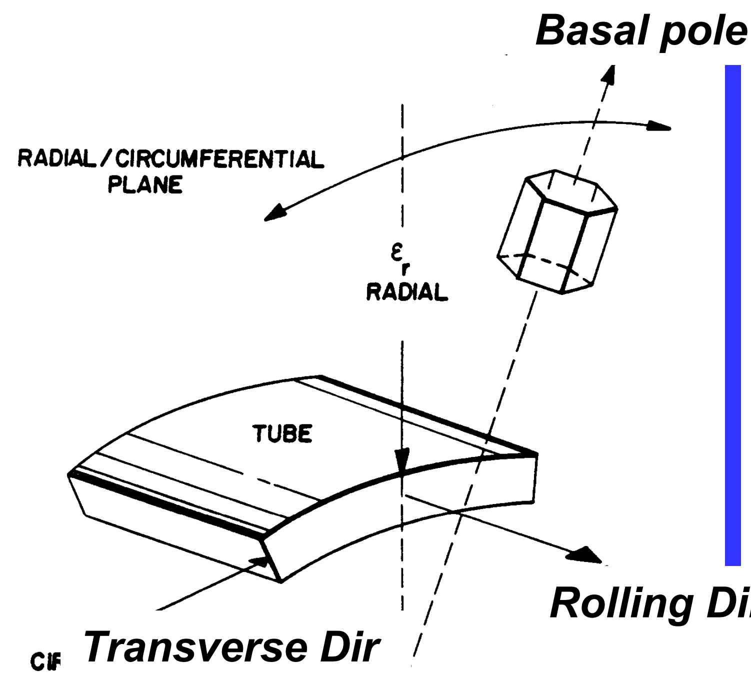


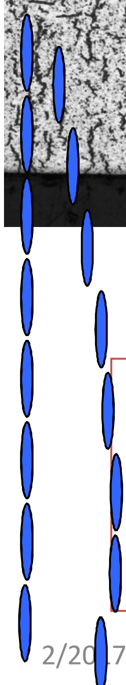
Without crystallographic texture

Individual hydride precipitates stack, rather than reorient, to give overall reorientation.









Precipitate reorientation was pictured as individual precipitates re-aligning, but reality is probably stacking.

Individual precipitates align themselves in the basal plane, but stack to reorient.

Conclusions

We understand & describe material behavior at the microstructural and atomistic scales

- **Grain growth, grain boundary mobility, annihilation, fission gas bubbles, pore distribution**
- **O/M ratio, defects, thermal conductivity, diffusion and diffusion pathways**

But, we model it on the continuum scale with all these features are homogenized

We have the models to simulate ceramics at the next level of refinement – mesoscale to capture the microstructure

- **Mesoscale models are sufficiently developed to simulate large scale problems**
- **Codes and computational capabilities can scale to large number of features**
- **Can couple to atomistic and continuum**
- **Enable understanding (and prediction) on this scale**
- **Enable designing and engineering materials at the meso-scale.**