# Hydride modeling approach for assessing the hydrid behavior in high burnup fuels

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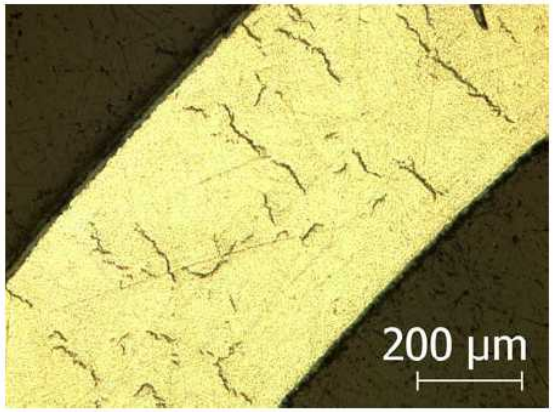
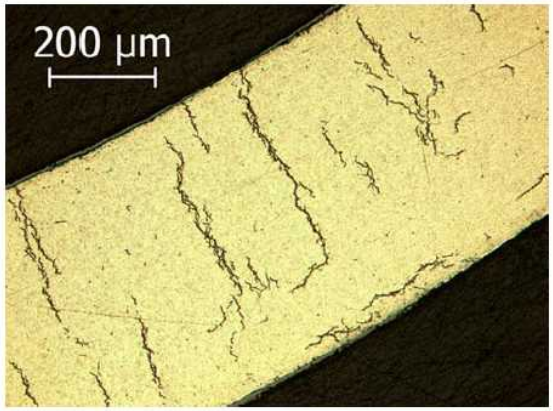



(a) 50% RHCF



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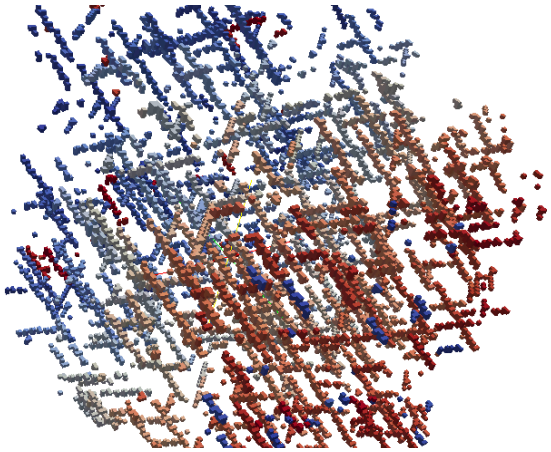
# Hydride modeling approach for assessing the hydride behavior in high burnup fuels

**Model objective: to simulate $\delta$-$ZrH_{1.5}$ formation at this microstructural scale.**

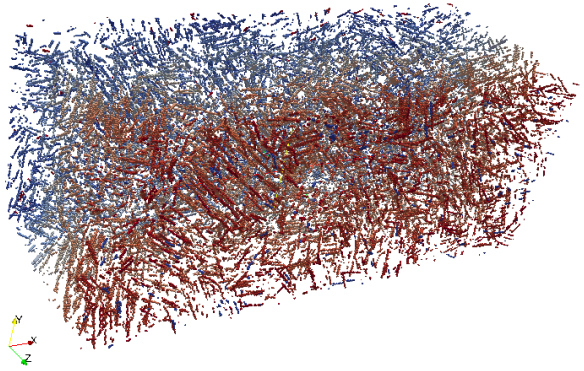
**Accomplishments: Developed a basic model to simulate precipitation and growth of $\delta$-$ZrH_{1.5}$ precipitates in the $\alpha$-Zr matrix**

- **In a Zircaloy-4 with correct grain and crystallographic texture**
- **Correct thermodynamic driving forces for the two phases**
- **With kinetic capability for nucleation and growth of precipitates.**

Bradbrook et al, JNM 1972



Precipitates in single crystal



Precipitates in polycrystalline

# Hybrid Potts-Phase Field Model
# Equation of State (Thermodynamics)

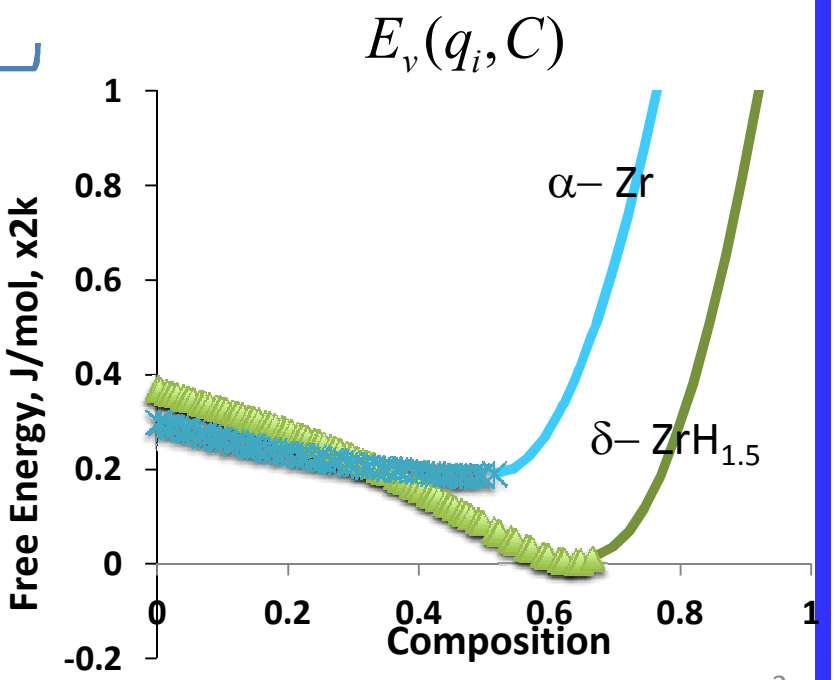
- **Combines elements of Potts and phase-field models to treat microstructure and composition evolution.**
- **EOS is a hybrid of volume free and interfacial energies obtained from both models.**

$$E_{hyb} = \sum_{i=1}^{N}\left(E_v(q_i,C) + \frac{1}{2}\sum_{j=1}^{n} J(q_i,q_j)\right) + E_{dC}$$

**Volume free energy** — **Interfacial free energy**

- $E_v(q_i,C)$ **is obtained from Thermo Cal**
- $E_{dC} = \int 2\kappa_C(\nabla C)^2 dV$

**M. Glazov, INL, 2013**



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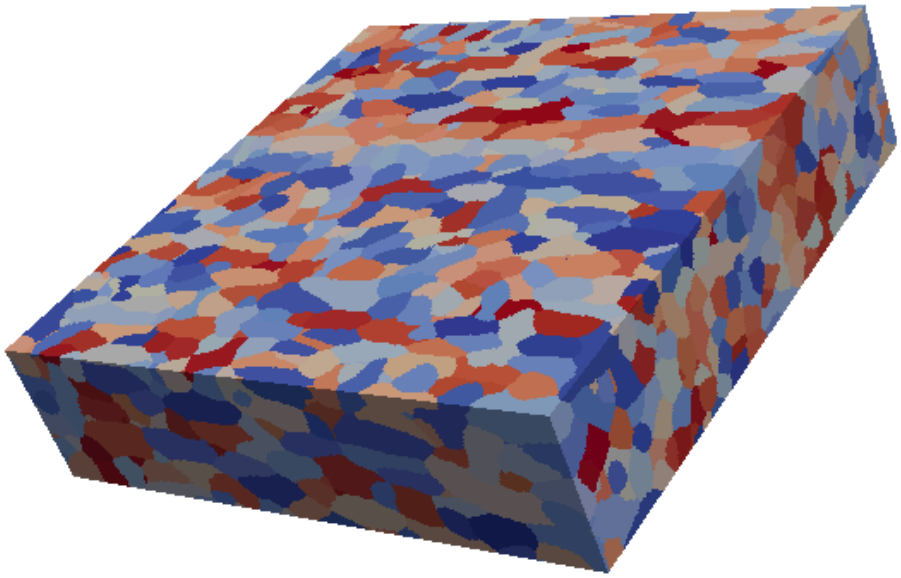
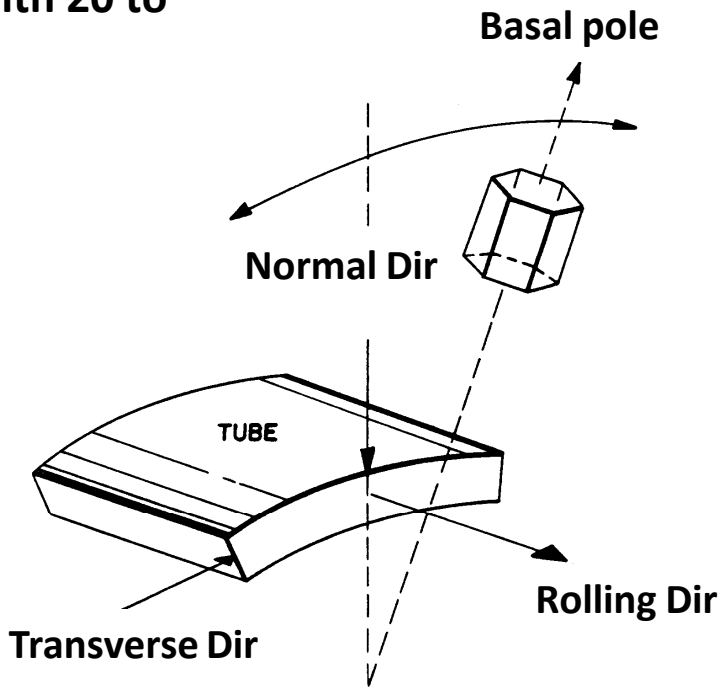
# Zircaloy-4 and other Zr-based claddings are known to have texture

**Pilgering process imparts texture**

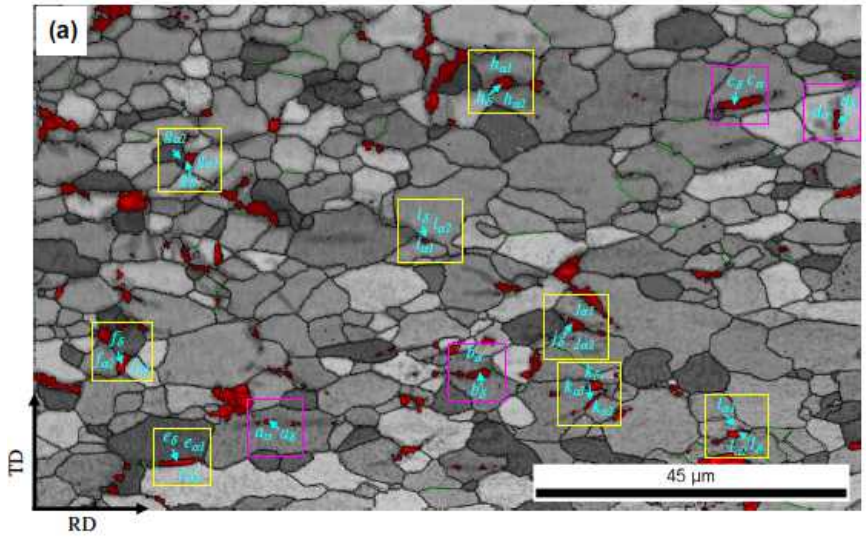
- **Grain shape, elongated ~2x in rolling direction**
- **Crystallographic, basal plane parallel to ND with 20 to 40$^{o}$ rotation around TD**

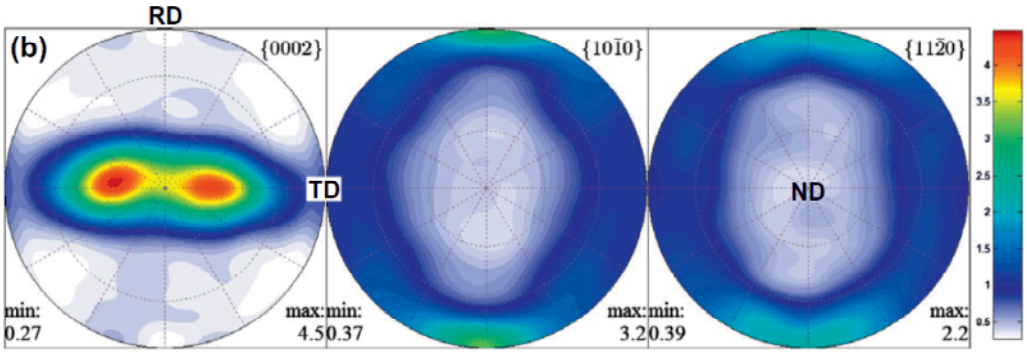


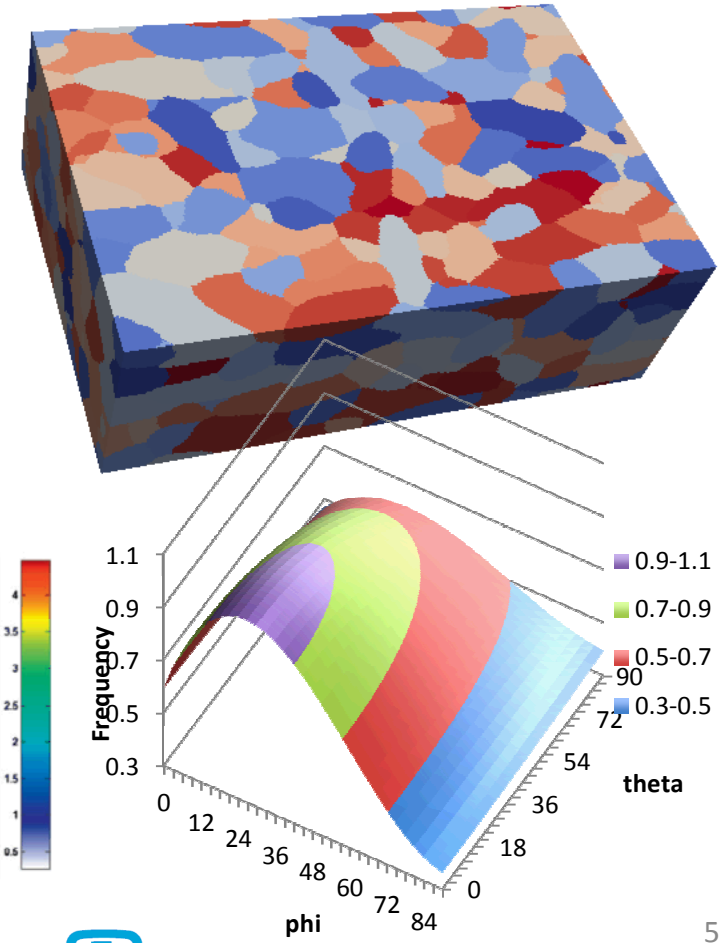


# Generated a Digital Zr-4 Microstructure

## from information available in the literature on grain and crystallographic texture



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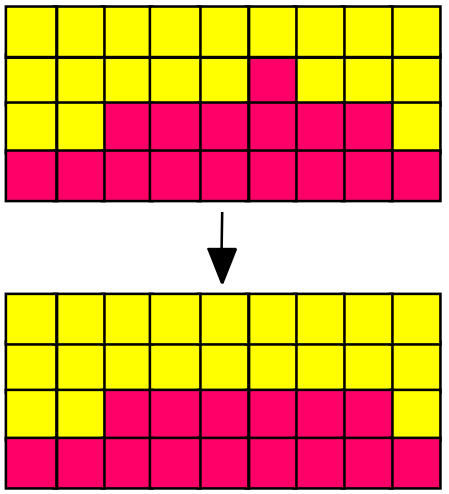
# Kinetic of Evolution Hybrid Model

- Microstructure is evolved in the same manner as Potts in response to local free energy using $E_{hyb}$
  - Metropolis algorithm
- Composition evolved as a phase field parameter.

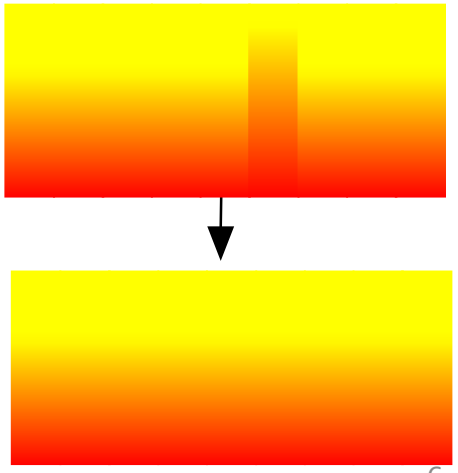
$$\frac{\partial C}{\partial t} = -M_c\left(\nabla^2 \frac{\partial E_v}{\partial C} - \kappa_c \nabla^4 C\right)$$

- Where $E_v$ is from the hybrid Free Energy

grain growth change pixel color



Composition change by diffusion



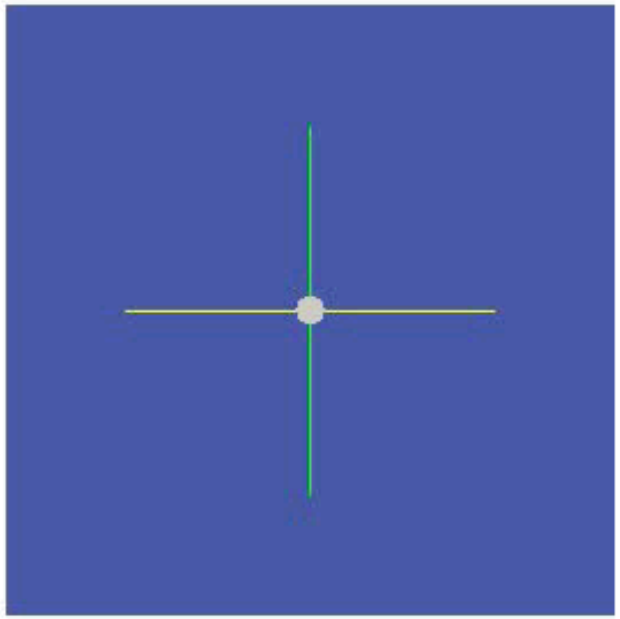
# Nucleation and growth of hydride precipitates is simulated by diffusion-controlled kinetics

**Model:**

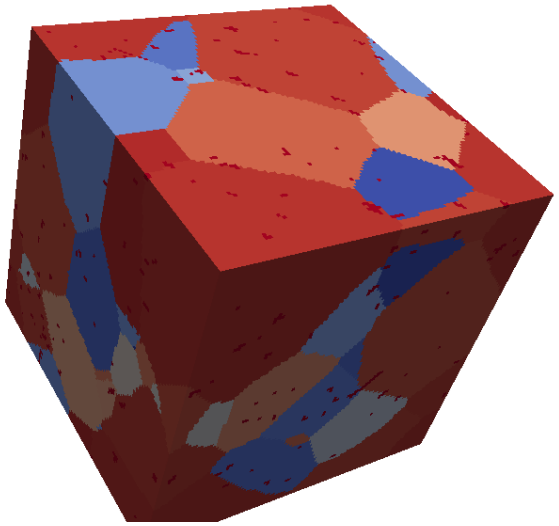
- **Nucleation sites are designated in the single crystal at random locations**
- **Nucleation rate is constant**
- **Energetic bias is given to growth direction of the lathes**

**Precipitate nucleation and growth**

**compositional evolution**

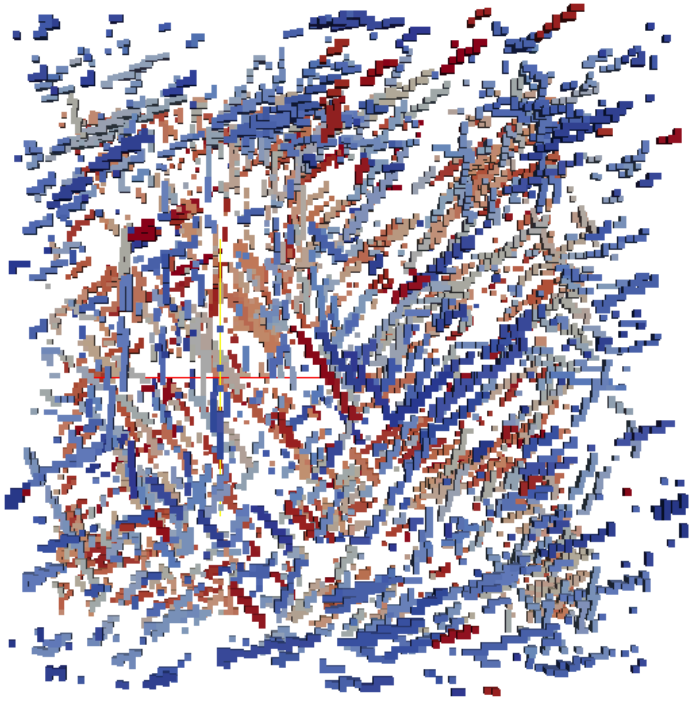


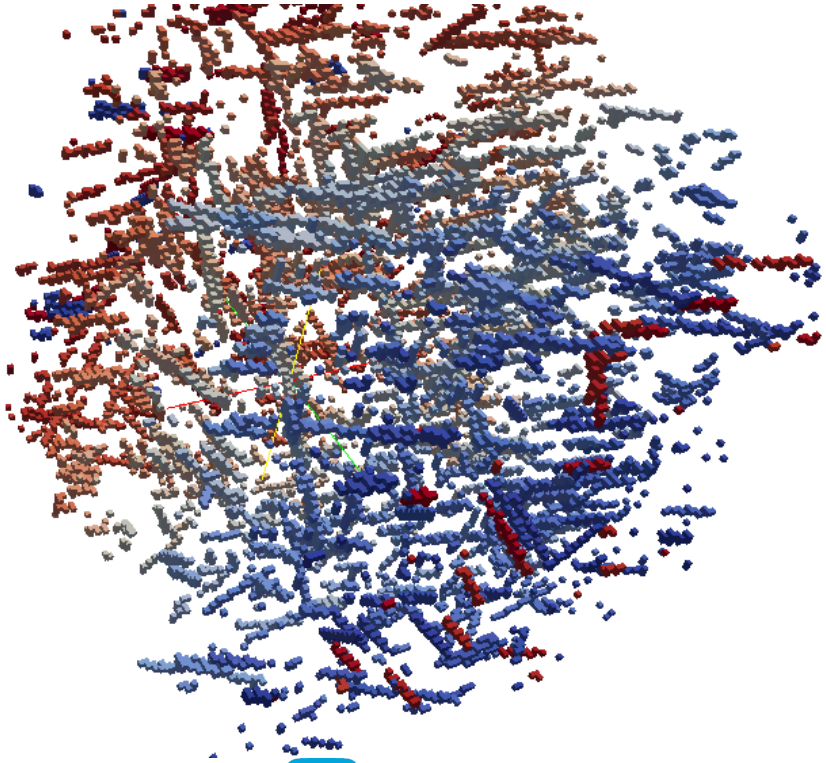
# $ZrH_{1.5}$ Precipitation Polycrystalline Zr



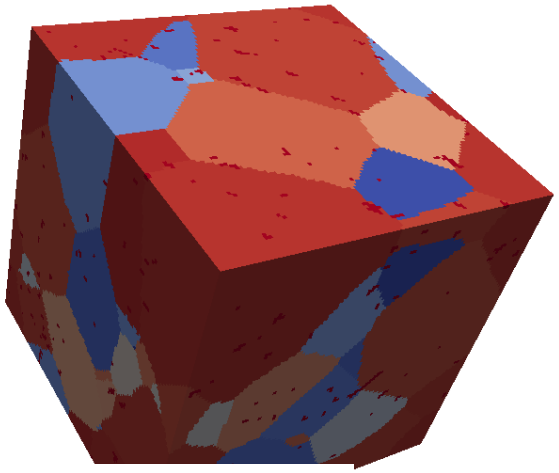
**Model:**

- **Nucleation at random locations and nucleation rate is constant**
- **Energetic bias is given to growth direction of the lathes with the orientation of the grain**



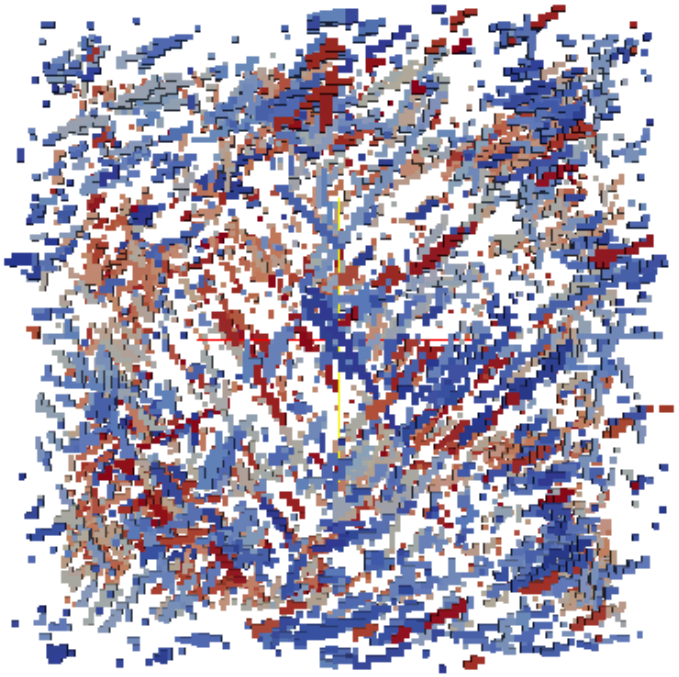


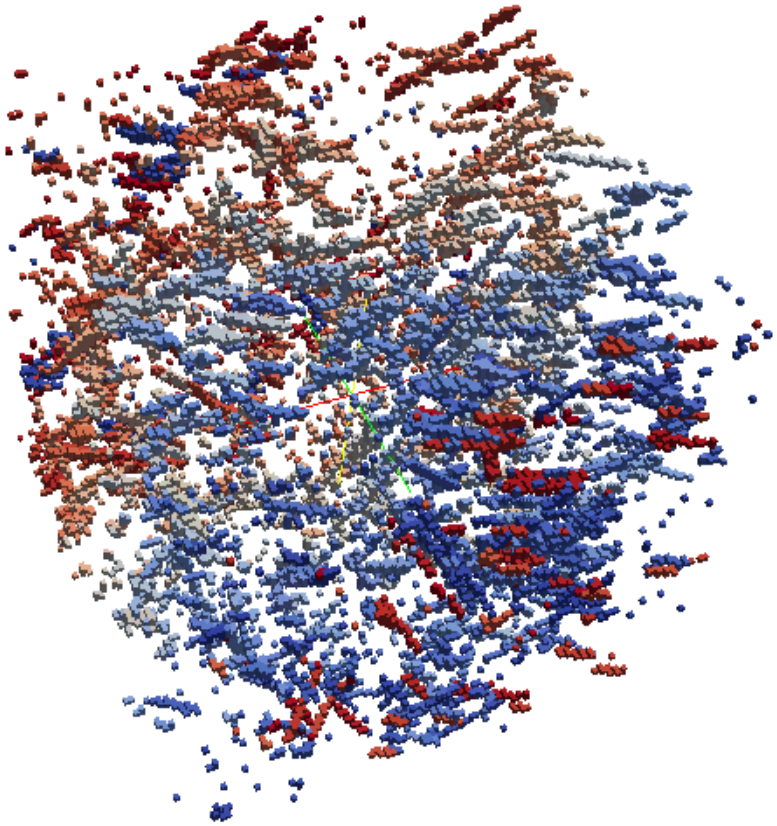
# $ZrH_{1.5}$ Precipitation Uniaxial Stress



**Model:**

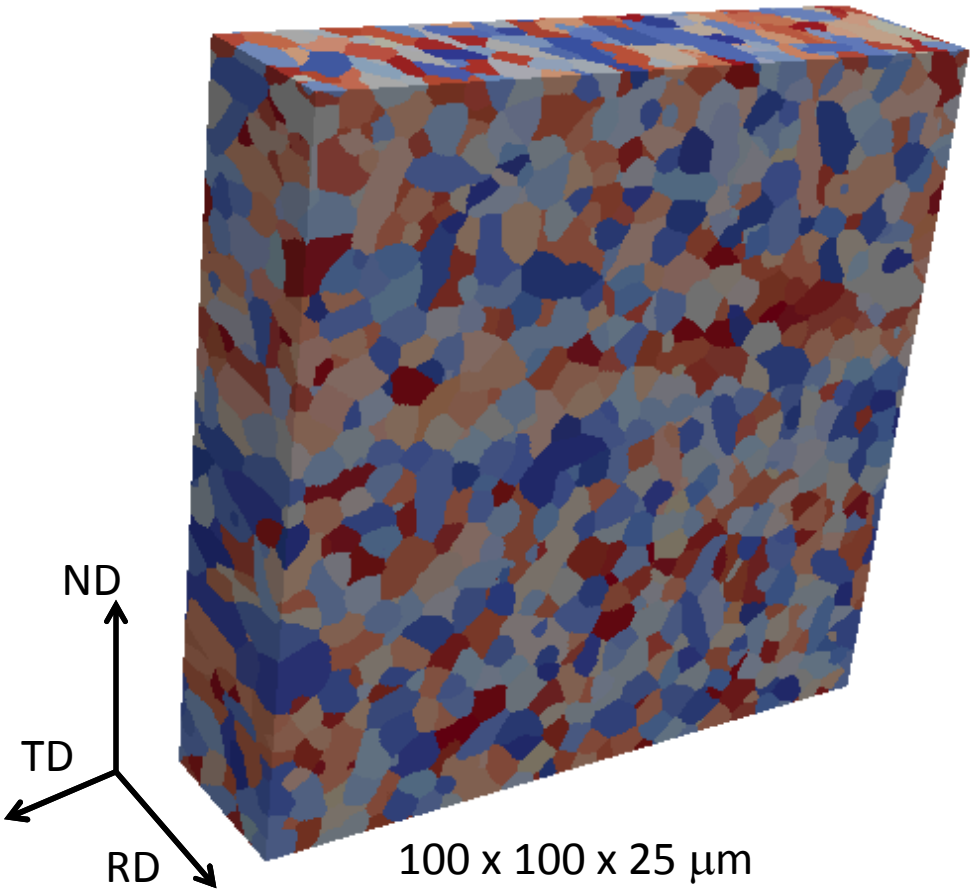
- **Same microstructure**
- **Energetic bias is given to growth direction of the lathes with the orientation of the grain and stress direction.**





# Nucleation and growth of hydride precipitates

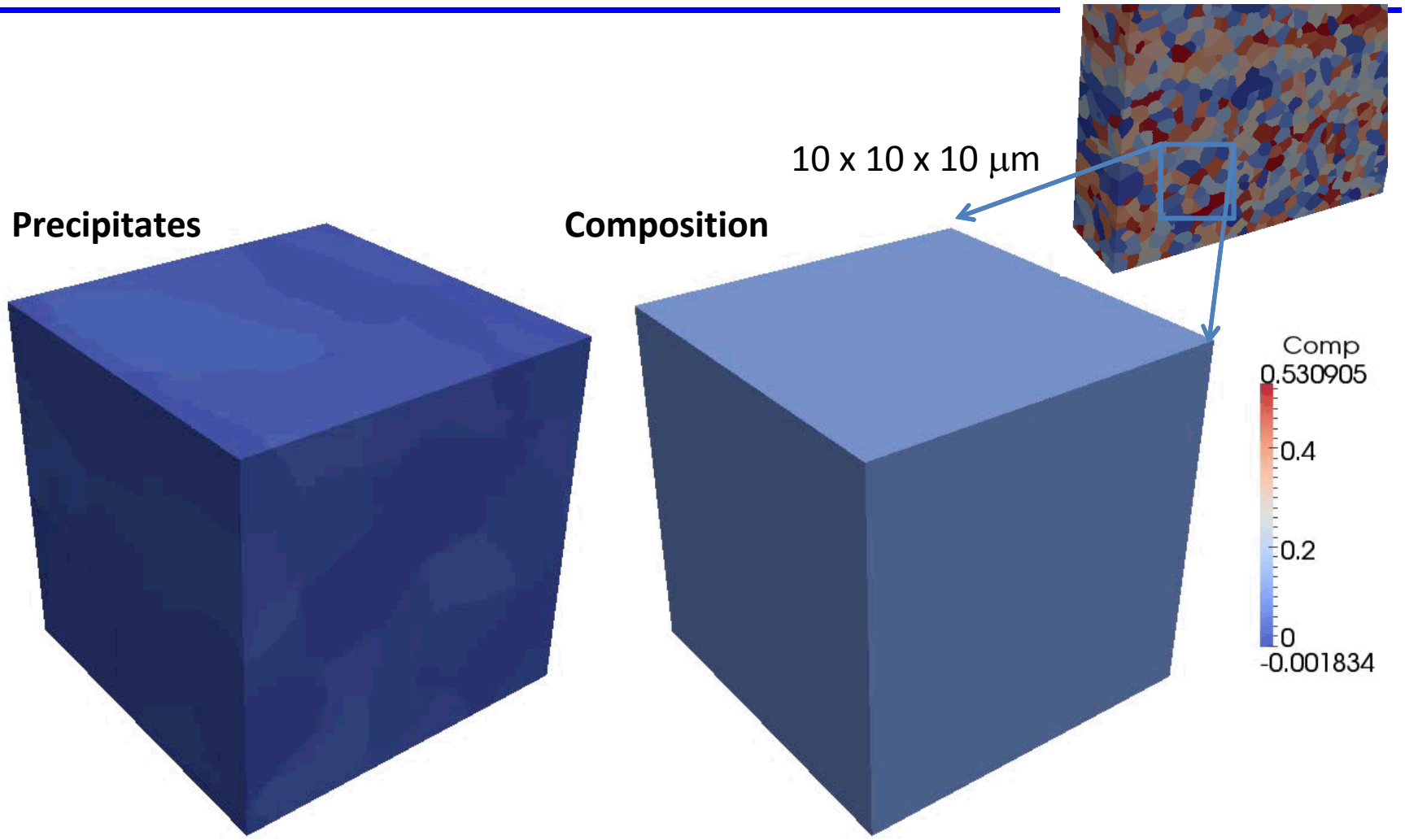
**Initial microstructure**




**Simulation:**

- **Microstructure as shown**
- **Basal plane alignment with RD**
- **1000 ppm H**
- **Zr-H thermo**
- **Nucleation and growth of precipitates**
- **Diffusion controlled kinetics**

# Nucleation and growth of hydride precipitates



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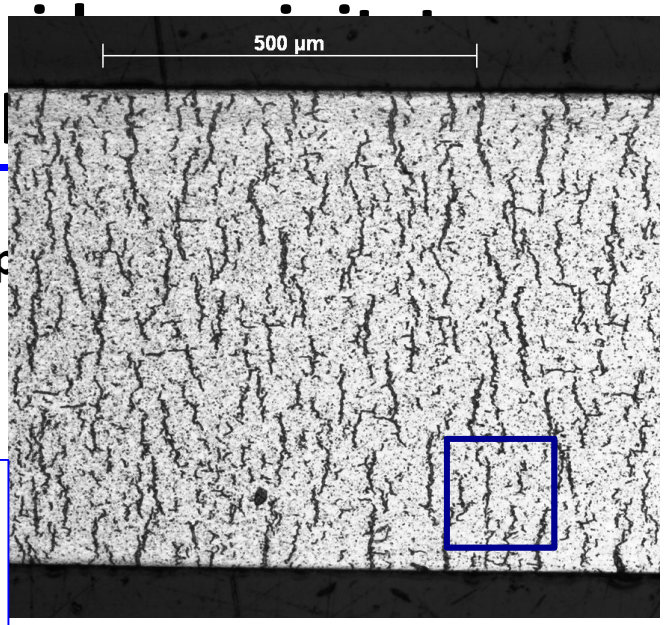
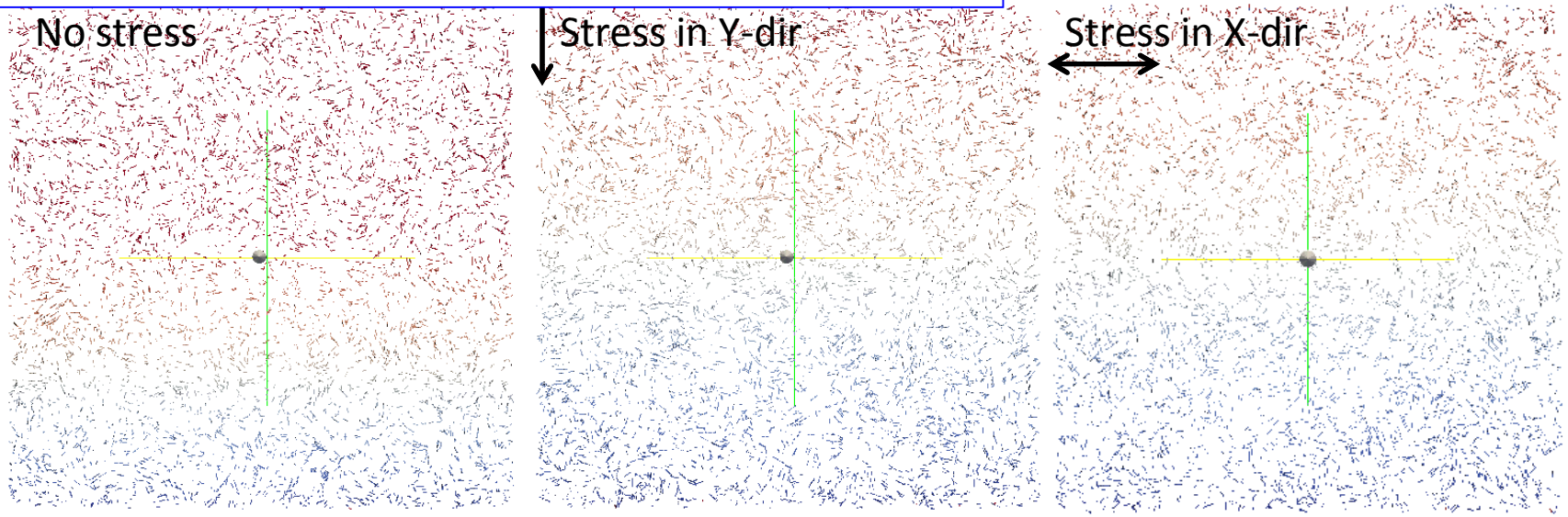
# Nucleation and growth of hyd
# Simulation resul




**Precipitates align themselves perpendicular to the app**

- **No stress, alignment is ~random along basal plane**
- **Stress along Y-dir, alignment is perpendicular**
- **Stress along Z-dir, alignment is *less perpendicular***

**However, simulated precipitates are uniformly distributed, individual precipitations do not re-align sufficiently to explain observed behavior**



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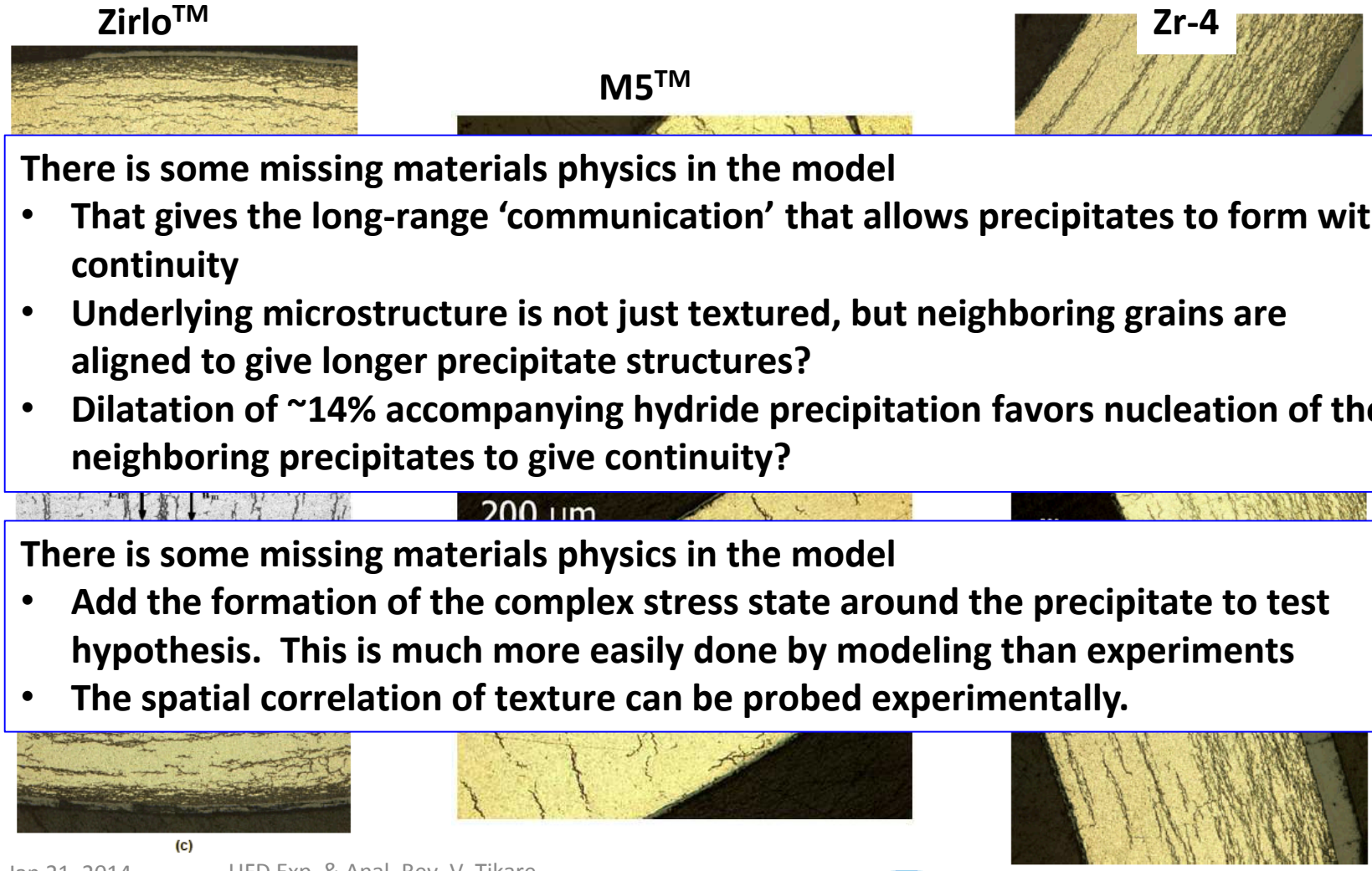
# All Zr-based claddings show long-range cooperative formation (Billone et al, ANL UFD report, 2013)




There is some missing materials physics in the model

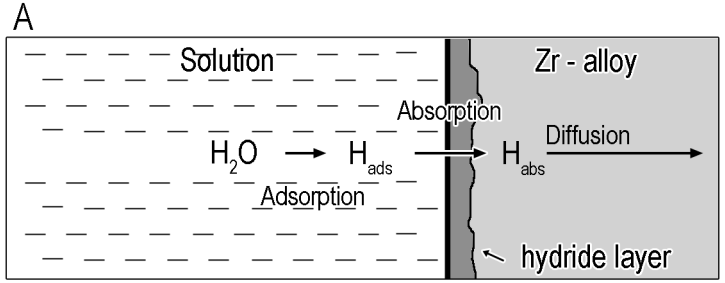
- That gives the long-range 'communication' that allows precipitates to form with continuity
- Underlying microstructure is not just textured, but neighboring grains are aligned to give longer precipitate structures?
- Dilatation of ~14% accompanying hydride precipitation favors nucleation of the neighboring precipitates to give continuity?

There is some missing materials physics in the model

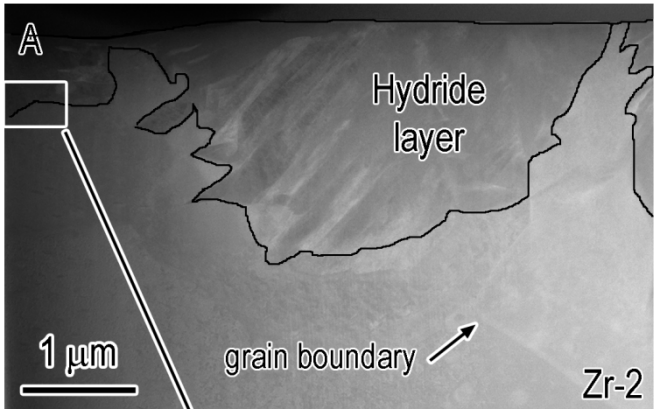
- Add the formation of the complex stress state around the precipitate to test hypothesis. This is much more easily done by modeling than experiments
- The spatial correlation of texture can be probed experimentally.

# First V & V Case: Surface Hydride Growth in Zr-Based Claddings (B. Clark et al, SNL, 2013)

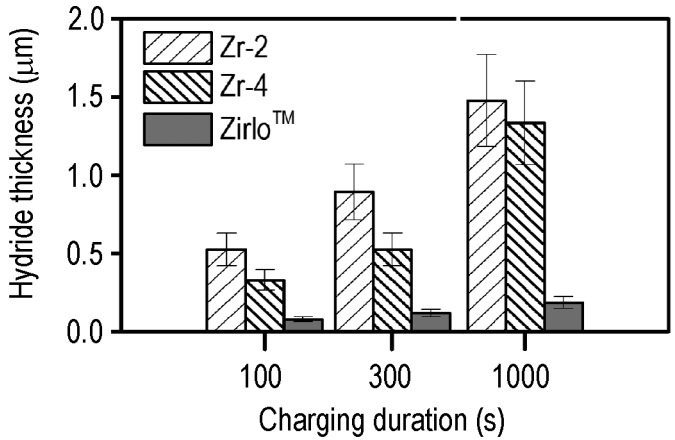
**Zr-based cladding were 'charged' with H**




**Large variation in hydride layer size from grain to grain.**




**Kinetics suggest diffusion controlled growth, $d \sim t^2$**



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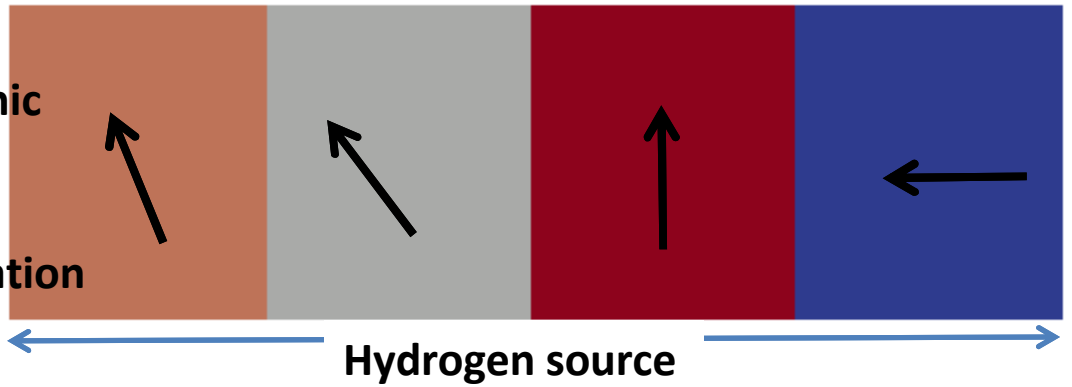
# First V & V Case: Surface Hydride Growth in Zr-Based Claddings (B. Clark et al, 2013)

**Simulation:**

- Underlying microstructure
- Arrows show crystallographic direction
- H source on surface
- Diffusion controlled, nucleation and growth of precipitates
- Variables
  - D of $\alpha$-Zr
  - D of $\delta$-$ZrH_{1.5}$




# First V & V Case: Surface Hydride Growth in Zr-Based Claddings (B. Clark et al, 2013)

**Interfacial growth of hydride precipitates cannot give the variation with these kinetics.**

**Which diffusion behaviors can lead to the variation in hydride growth that is grain dependent?**

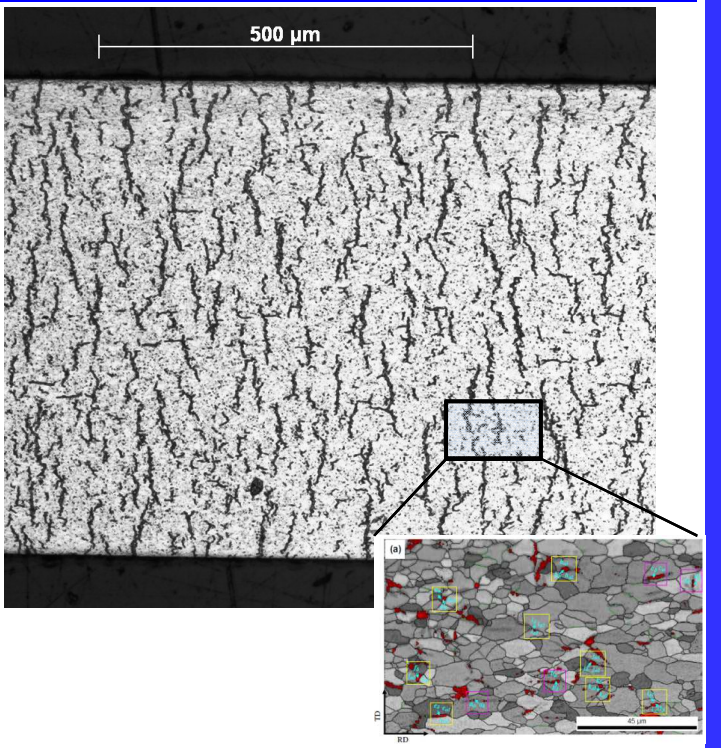
- **D is a function of crystallographically direction in $\alpha$-Zr**
- **Kearns reported ~$D_a = 2D_c$, not enough to give variation in precipitate growth.**
- **Tried $D_a = 100D_c$, still no variation**

- **D is a function of crystallographically direction in $\delta$-$ZrH_{1.5}$**
- **Need $D_a \sim 100D_c$, to get the differential growth.**

Kearns, JNM 1972

# Experimental Data Needed

- Microscopy
  - SEM of the hydrides
  - What are we seeing?
  - How do the individual precipitates align
  - Microstructure of $\alpha$–Zr
- Compositional effects
  - Nb and other alloying elements
  - Free energy calculations




W. Qin, JNM , 2011

# Hydride Roadmap
# Where are we and where are we going?

- FY13: demonstrate basic hydride formation in Zr-4 microstructure
- Have a model:
  - Realistic microstructure
  - Reasonably large scale (can simulate 100's of grains.)
  - Actual thermodynamics (free energies)
  - Hydride formation (nucleation and growth)
  - Kinetics are diffusion controlled
  - Can capture radiation effects
    - Nucleation sites and rates
    - Diffusion

# Hydride Roadmap
# Where are we and where are we going?

- **FY14: Further develop the model to simulate hydride formation**
  - **More complex chemical compositions**
  - **V&V exercises**
  - **Incorporating model into MOOSE/BISON/MARMOT**
  - **Adding temperature effects, both transient and non-isothermal**
  - **Add micro-mechanical coupling to**
    - **capture the local stress state around hydride precipitates**
    - **Influence hydride nucleation and growth**

**Need the coupling to the micro-mechanical model to address the long-range cooperative hydride precipitate alignment.**