Simulation of Hydride Formation in Zr-Based Claddings during Dry Storage

SAND2014-0657C

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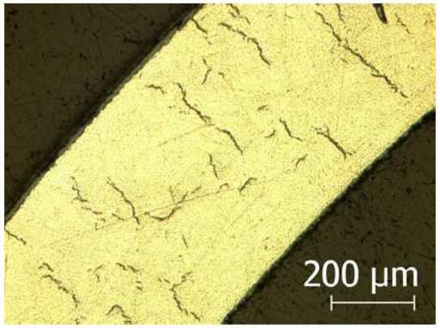
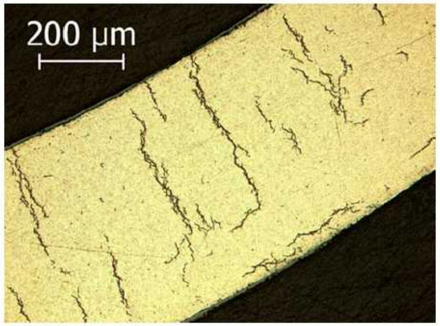


(a) 50% RHCF



Billone et al, ANL, 2013

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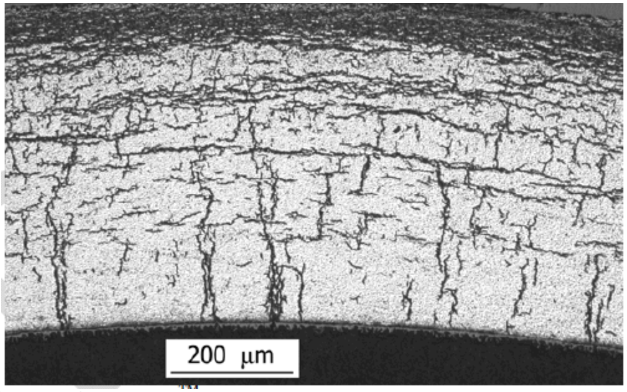
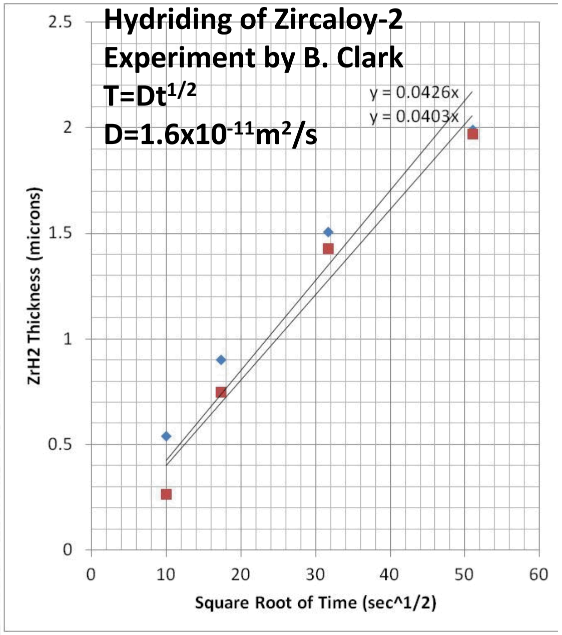
ZrH_2 Formation during Dry Storage May Limit the Lifetime of Used Nuclear Fuel Cladding

Hydride formation in Zr-based cladding is known to degrade clad reliability during during dry storage

- **The drying cycle dissolves the ZrH_2.**
- **During dry storage as the clad temperature decreases, the ZrH_2 precipitates on grain boundaries, dislocation loops and, possibly, other defects.**
- **ZrH_2 forms perpendicular to the tensile stress**
- **It morphology appears to be needle-like lathes**
- **B. Clark's experiments indicate the ZrH_2 growth is diffusion controlled**
- **Microstructure, stress, composition, H pick-up, temperature are know to Control ZrH_2 formation & morphology.**

Zirlo 720 wppmH 140 MPa hoop stress 400 °C

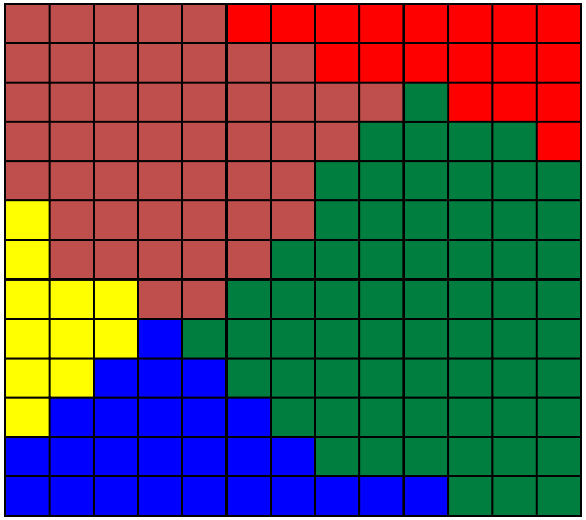



Hydriding of Zircaloy-2
Experiment by B. Clark
$T=Dt^{1/2}$
$D=1.6x10^{-11}m^2/s$

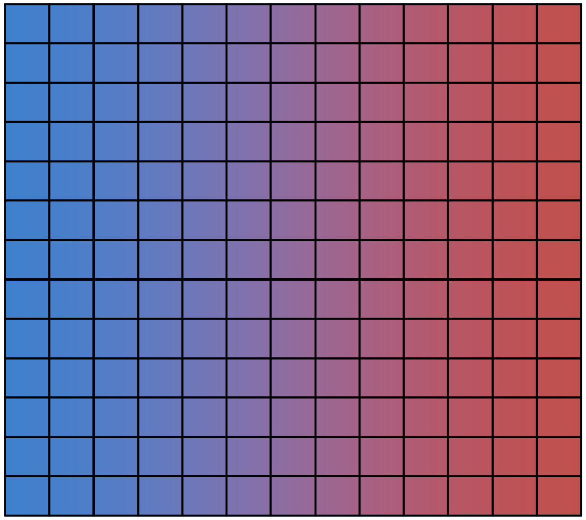
Representation of Microstructure and Composition Hybrid Model

- **Potts kMC digitizes represents microstructure using spins q_i**
- **Phase field represents composition with field variable C_i**
  - **Both are on the same grid**

Microstructure



Composition



Equation of State (Thermodynamics) Hybrid Model

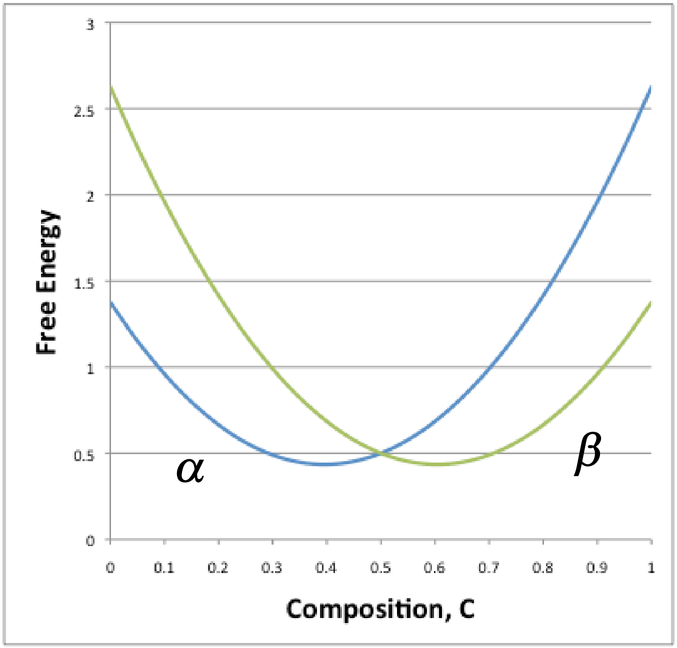
- **EOS is a function of volume free energy and interfacial energies.**

$$E_{hyb} = \sum_{i=1}^{N} \underbrace{\left(E_v(q_i, C) \right.}_{\text{Volume free energy}} + \underbrace{\left. \frac{1}{2} \sum_{j=1}^{n} J(q_i, q_j) \right) + E_{dC}}_{\text{Interfacial free energy}}$$

$$E_{dC} = \int 2\kappa_C (\nabla C)^2 dV$$

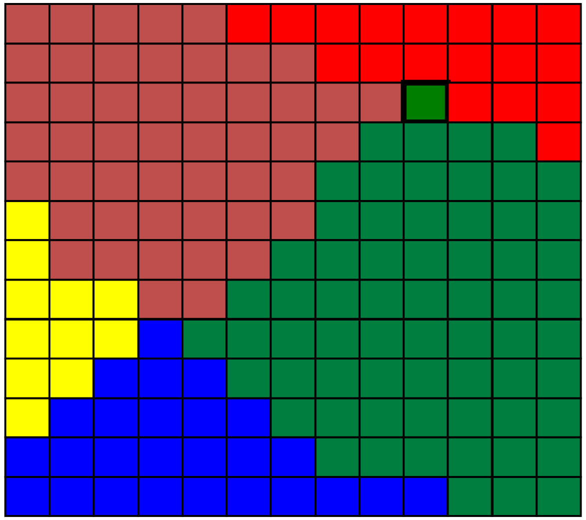
An example of E_v

$$E_v = a\left[(C - C_1)^2 + (C_2 - C)^2 \right] + b\left[(C - C_3) q_\alpha + (C_4 - C) q_\beta \right]$$



Representation of Microstructure and Composition kMC

- **Potts kMC digitizes space into discrete 'bits' of material**
  - **An ensemble of particles populate the lattice**
  - **Each color can represent a membership in a phase and / or feature (i.e. grain)**
  - **Each color can also represent composition, but true gradients in composition would require huge simulations**



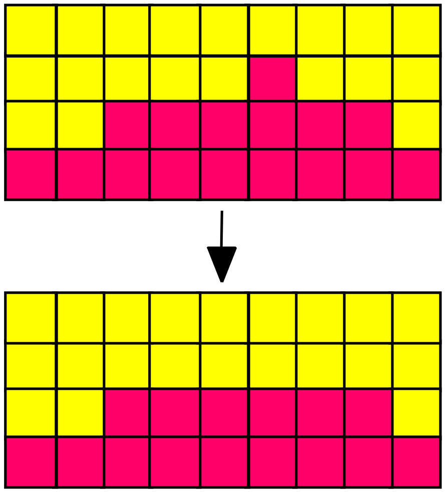
Kinetic of Evolution Hybrid Model

- 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.

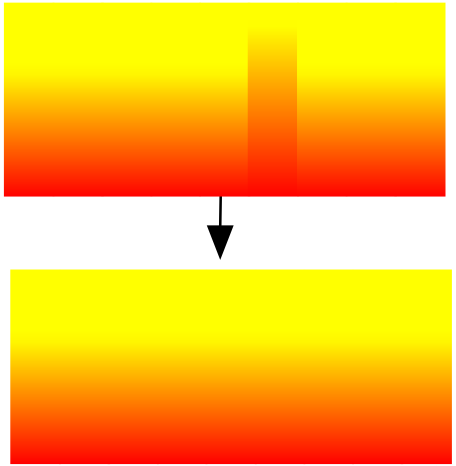
$$\frac{\partial C}{\partial t} = -M_c\left(\nabla^2 \frac{\partial E_v}{\partial C} - \kappa_c \nabla^4 C\right)$$

- Where E_v is from the hybrid Free Energy

grain growth change pixel color

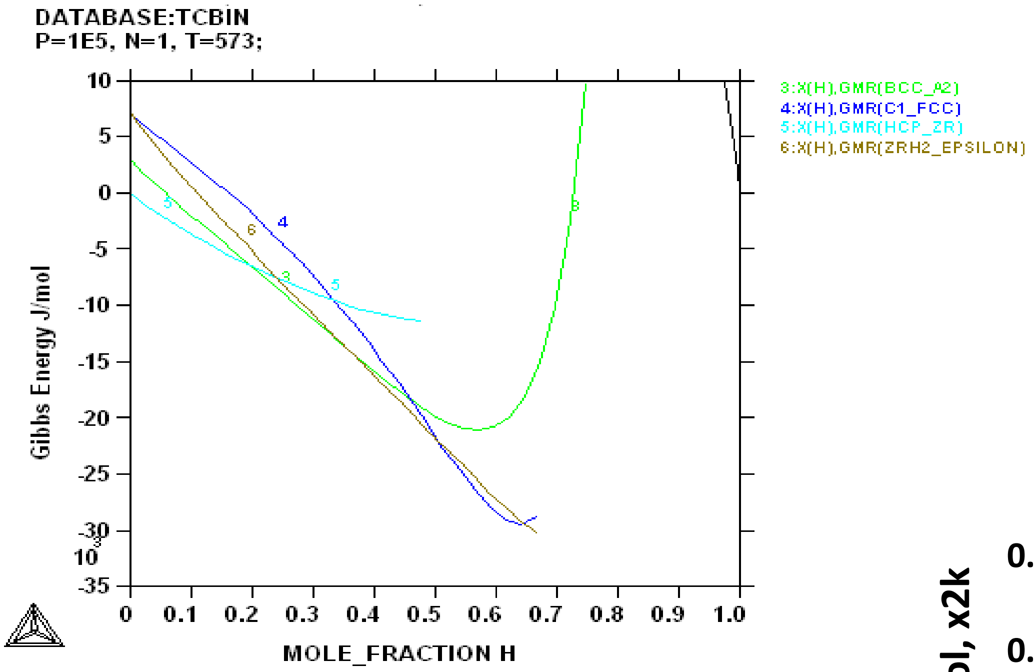


Composition change by diffusion

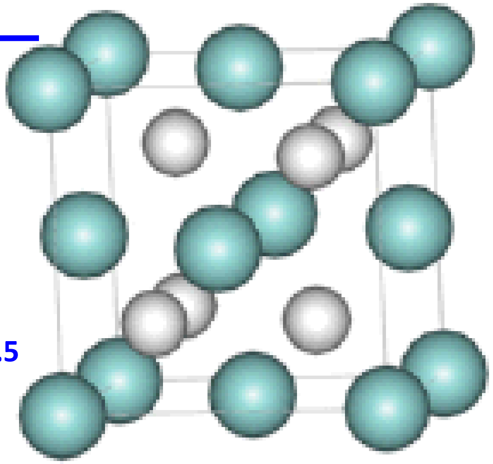


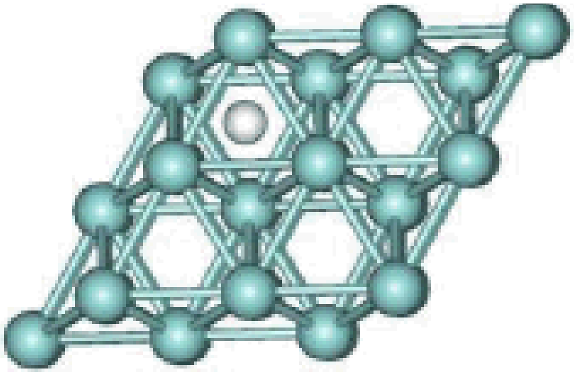
Phase Equilibrium Calculations for Zr and $ZrH_{1.5}$

M. Glazoff, INL

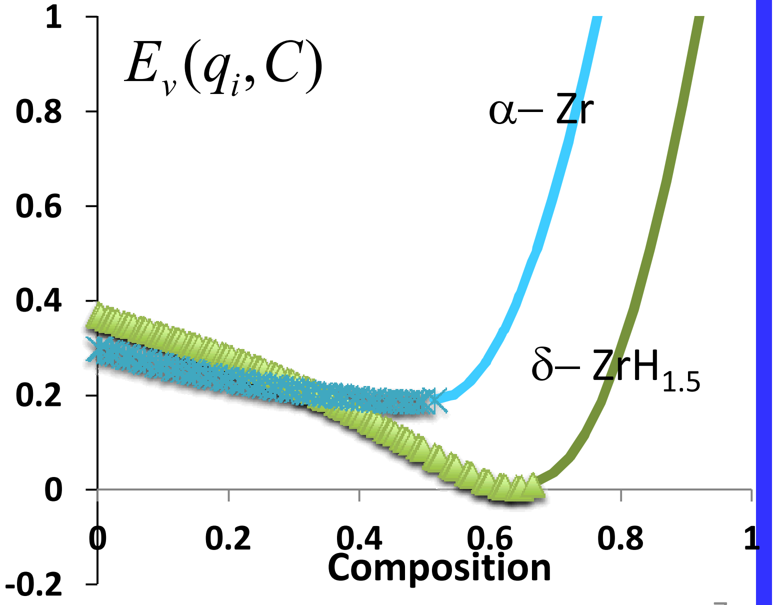


FCC $ZH_{1.5}$





HCP Zr



Morphology of $ZrH_{1.5}$ Precipitates

$ZrH_{1.5}$ Precipitates

- Intragranular
- needle-like lathes
- Cubic (FCC)
- specific crystallographic orientation with HCP Zr
- Growth is by nucleation and diffusion-controlled growth

Hydride Precipitates orient on $(\mathbf{111})_\delta$ $//(0001)_\alpha$; $[1\bar{1}0]_\delta//[\mathbf{11\bar{2}0}]_\alpha$

$(\mathbf{111})_\delta$ $//(10\bar{1}0)_\alpha$; $[1\bar{1}0]_\delta//[\mathbf{11\bar{2}0}]_\alpha$

Bradbrook et al, JNM 1972

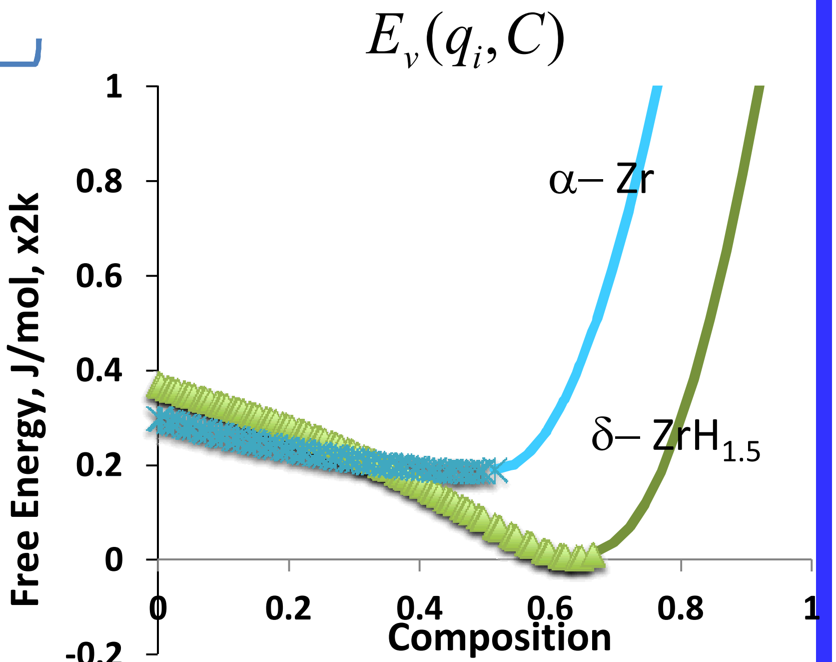
Hybrid Potts-Phase Field Model
Equation of State (Thermodynamics)

- **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.**

$$E_{hyb} = \sum_{i=1}^{N}\left(\underbrace{E_v(q_i,C)}_{\text{Volume free energy}} + \underbrace{\frac{1}{2}\sum_{j=1}^{n} J(q_i,q_j)\Big) + E_{dC}}_{\text{Interfacial free energy}}\right.$$

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

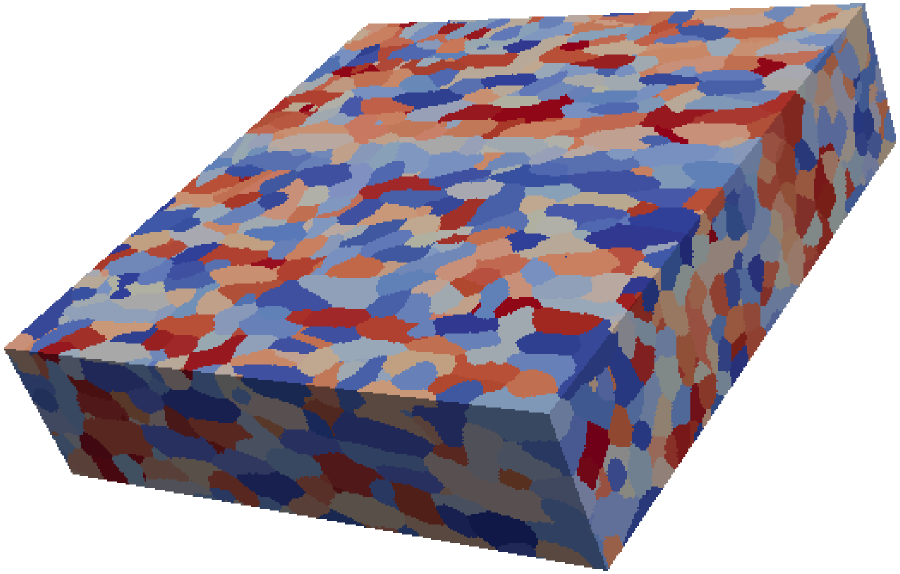
M. Glazov, INL, 2013

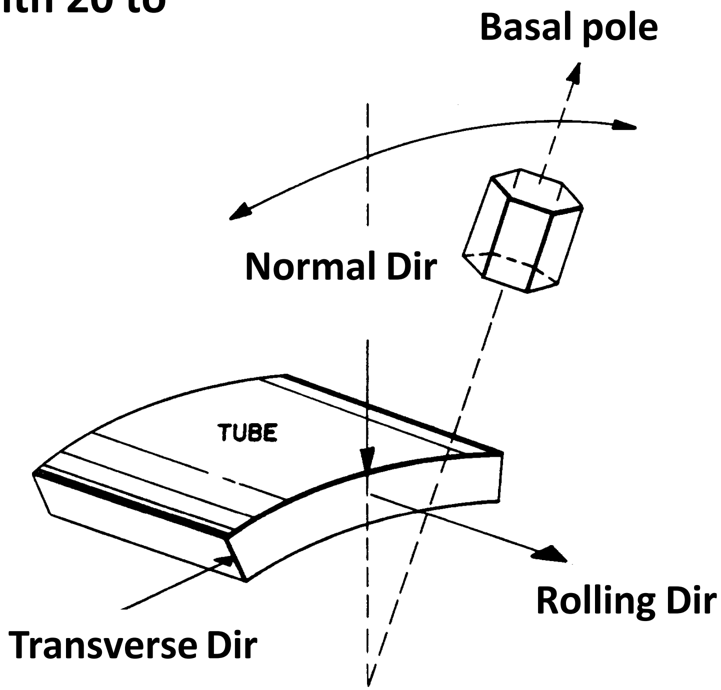


Zircaloy-4 and other Zr-based claddings are known to have texture

Pilgering process imparts texture

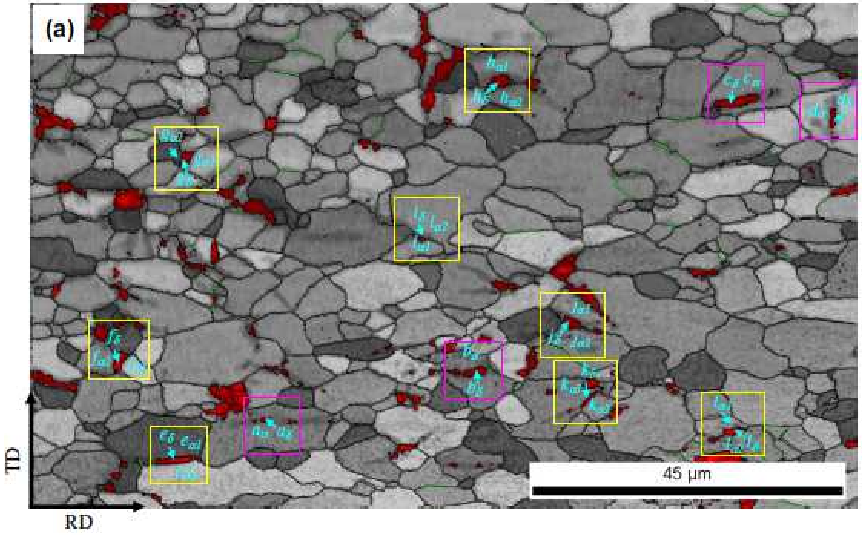
- Grain shape, elongated ~2x in rolling direction
- Crystallographic, basal plane parallel to ND with 20 to 40^{o} rotation around TD

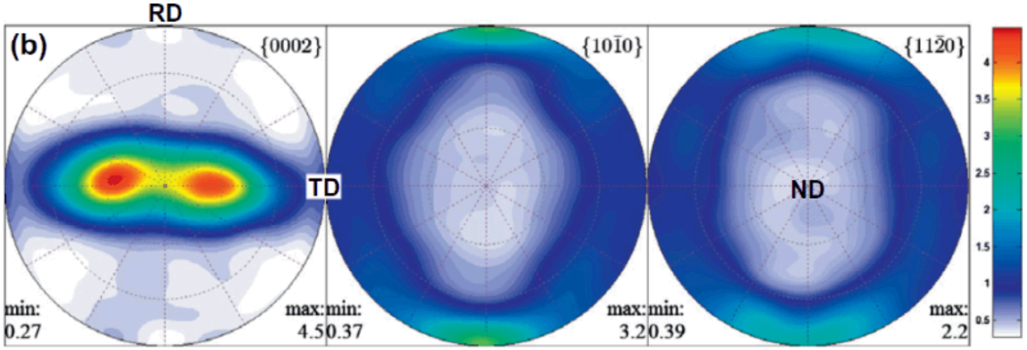


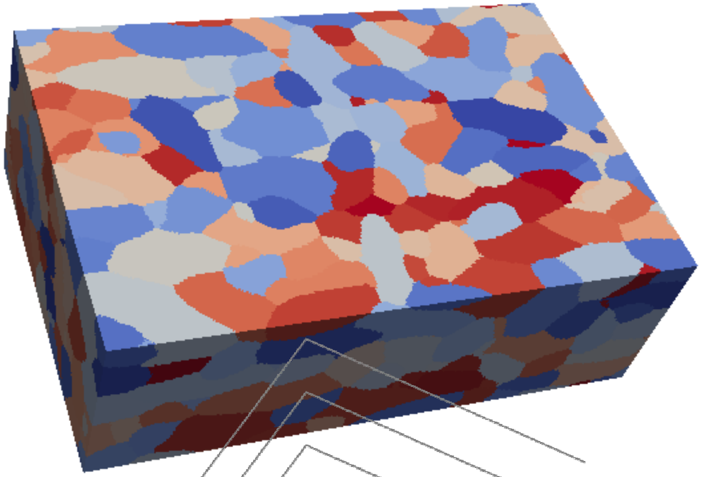


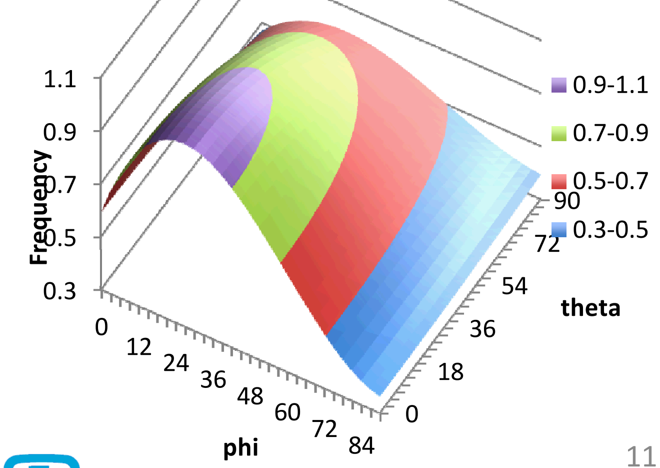
Generated a Digital Zr-4 Microstructure

from information available in the literature on grain and crystallographic texture









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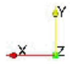
Nucleation and growth of hydride precipitates is simulated by diffusion-controlled kinetics

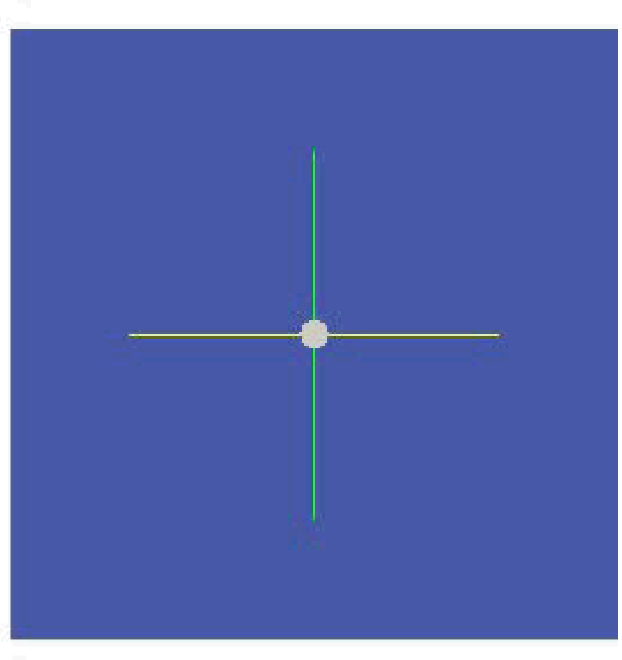
Model:

- 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

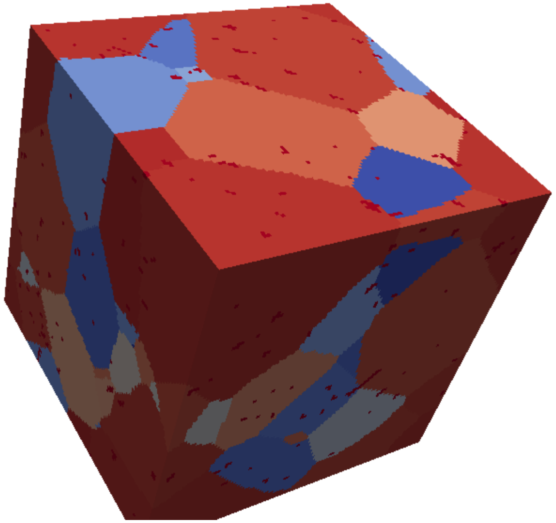
Precipitate nucleation and growth



compositional evolution

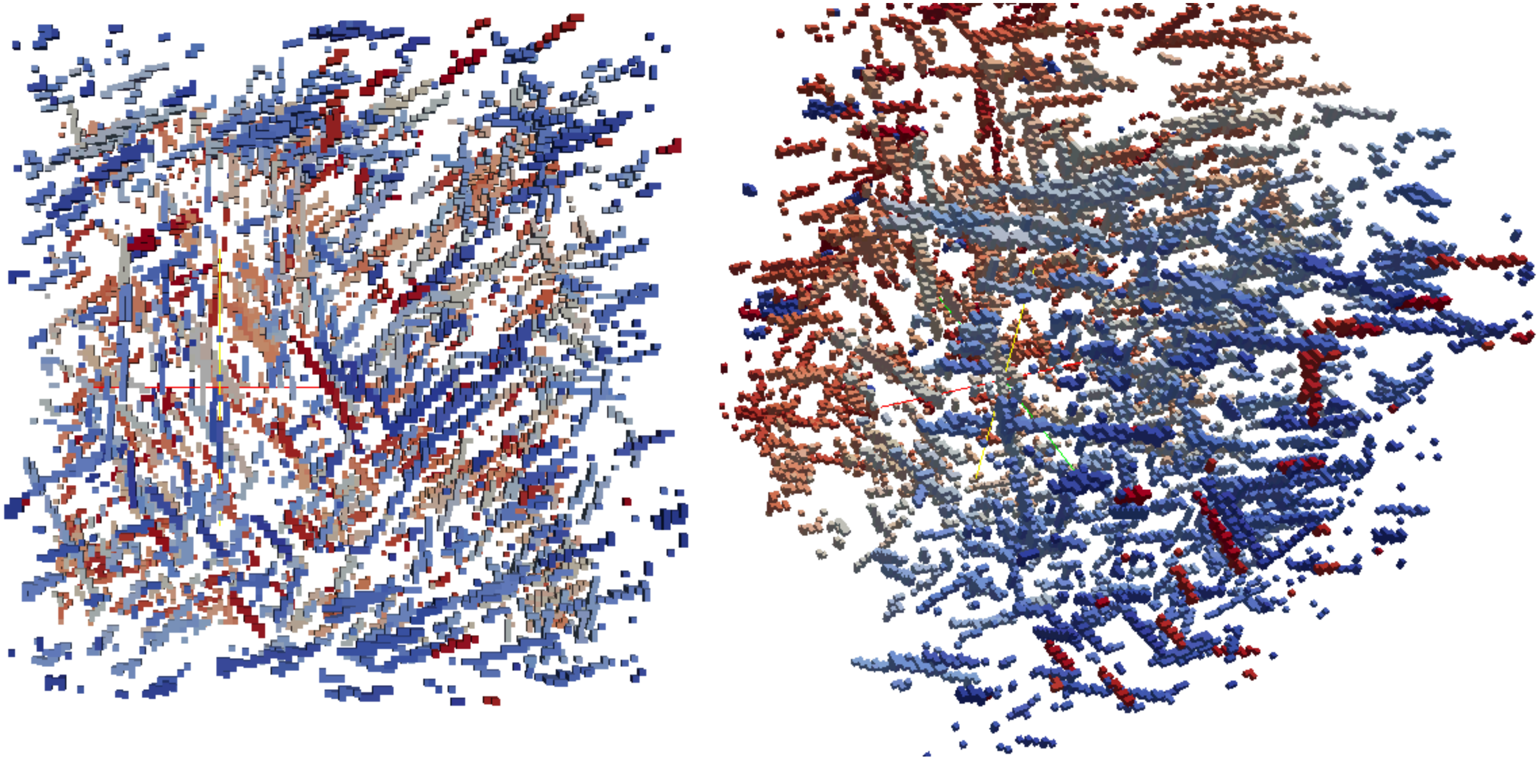


$ZrH_{1.5}$ Precipitation
Polycrystalline Zr

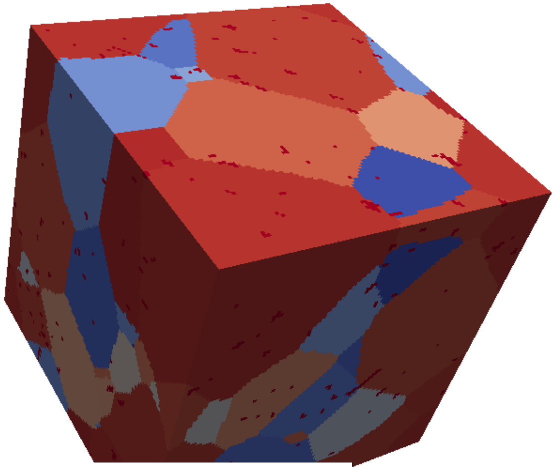


Model:

- **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**

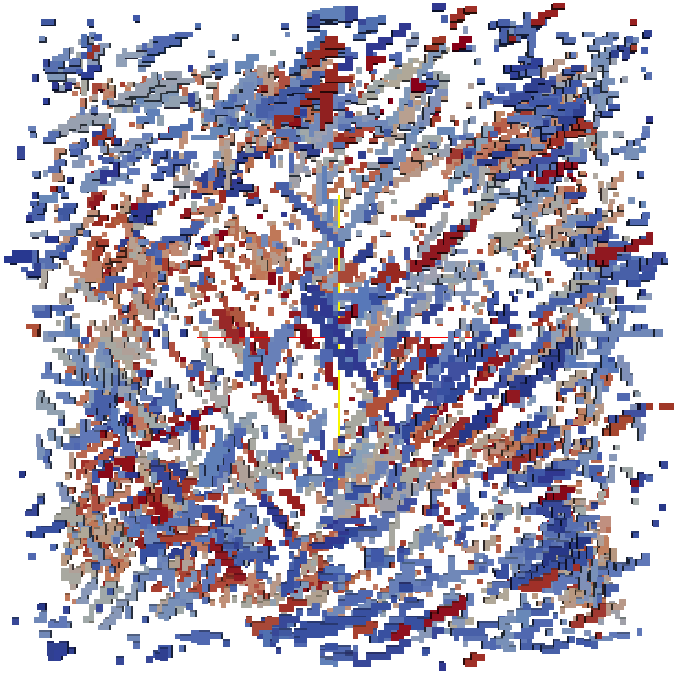


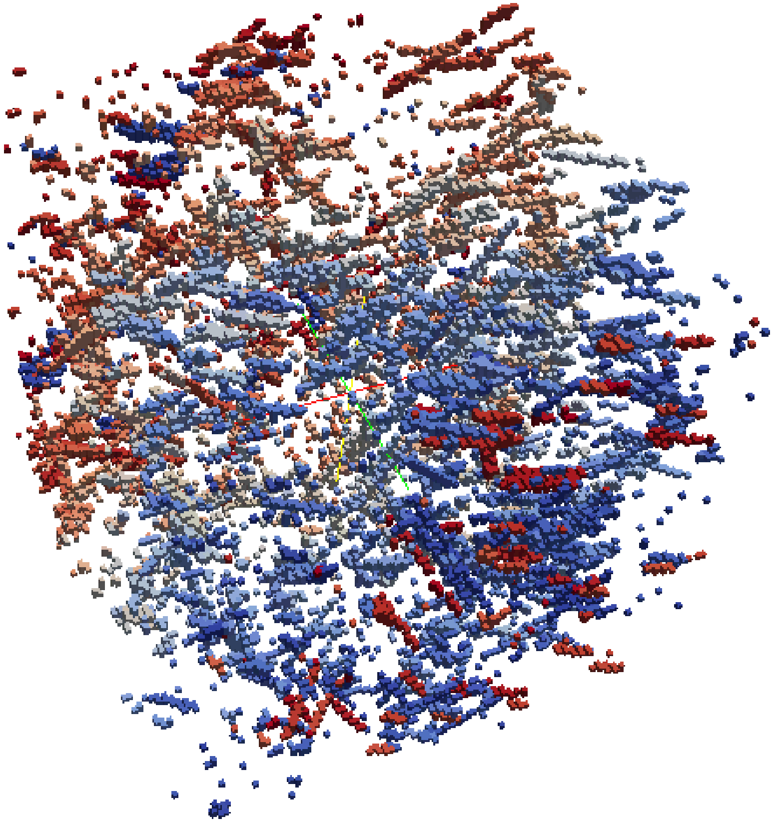
$ZrH_{1.5}$ Precipitation
Uniaxial Stress



Model:

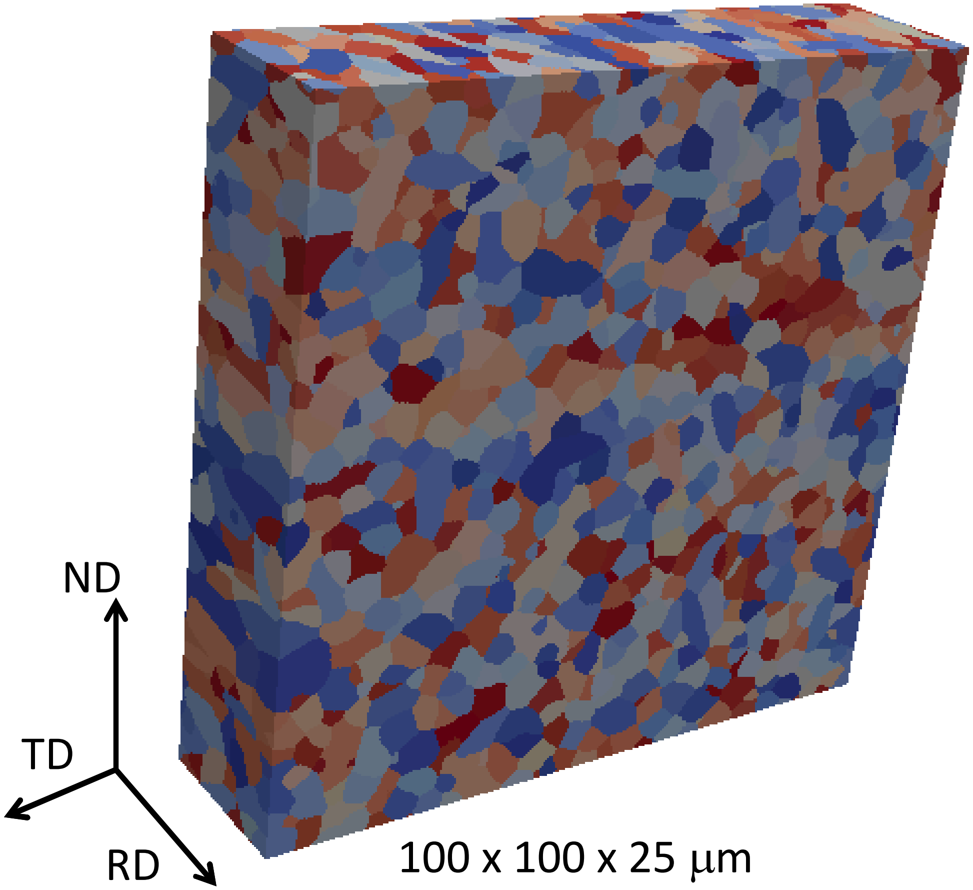
- **Same microstructure**
- **Energetic bias is given to growth direction of the lathes with the orientation of the grain and stress direction.**





Nucleation and growth of hydride precipitates

Initial microstructure

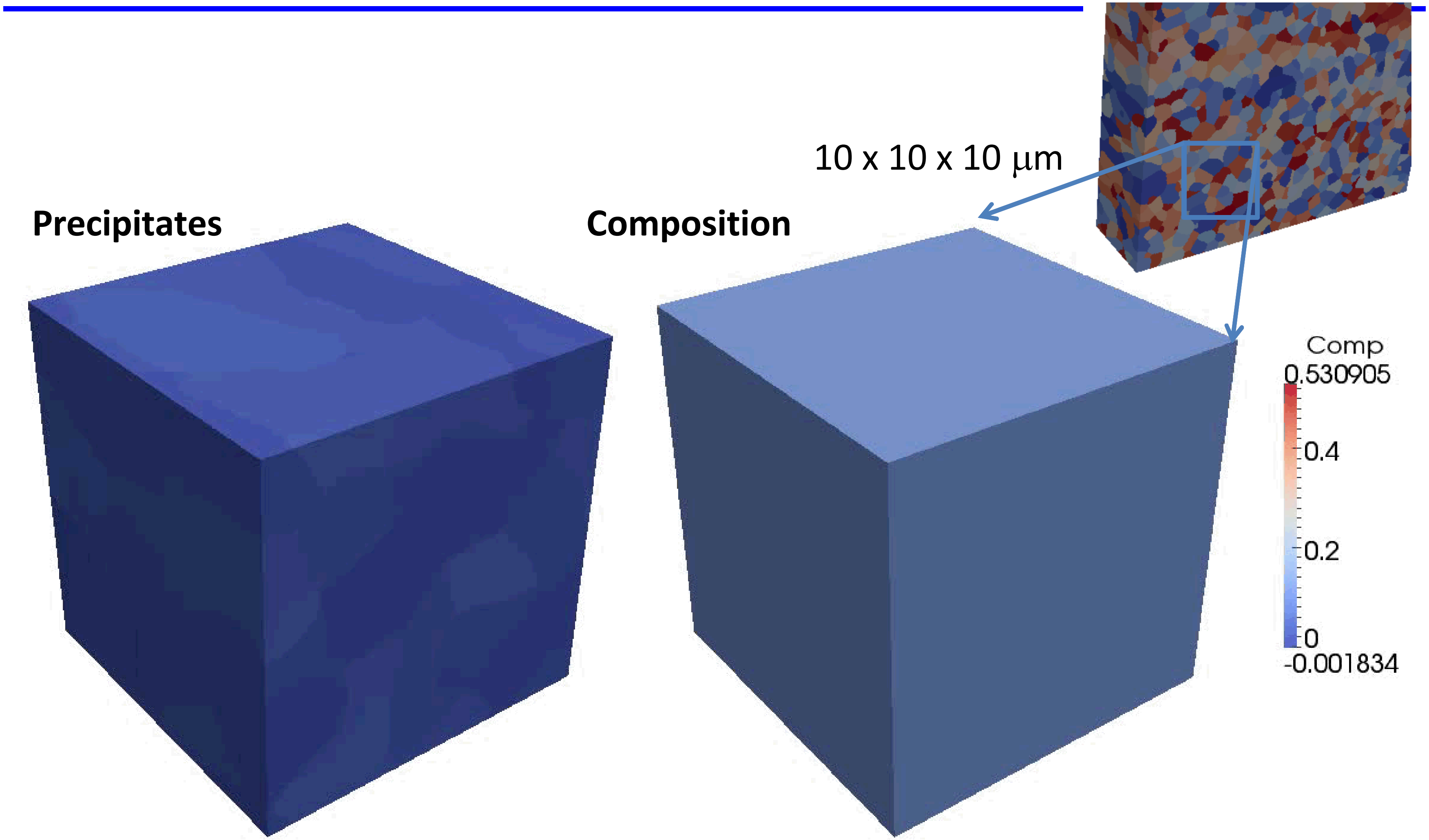


100 x 100 x 25 μm

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

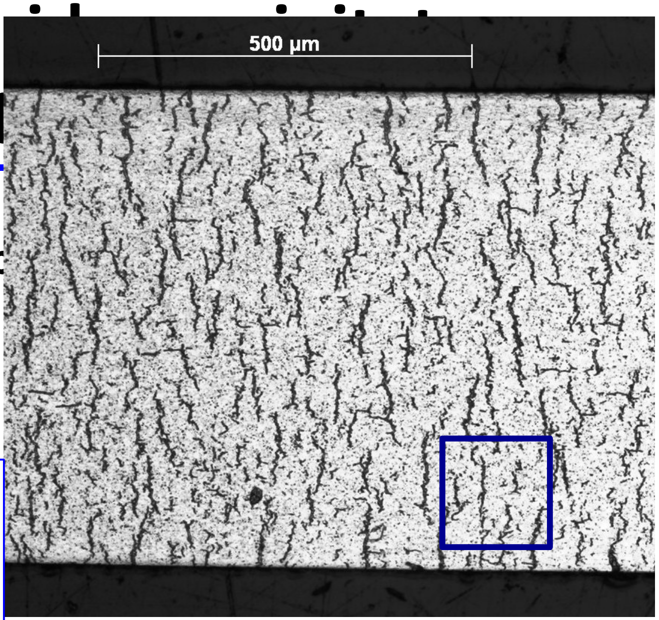


Precipitates

Composition

10 x 10 x 10 μm

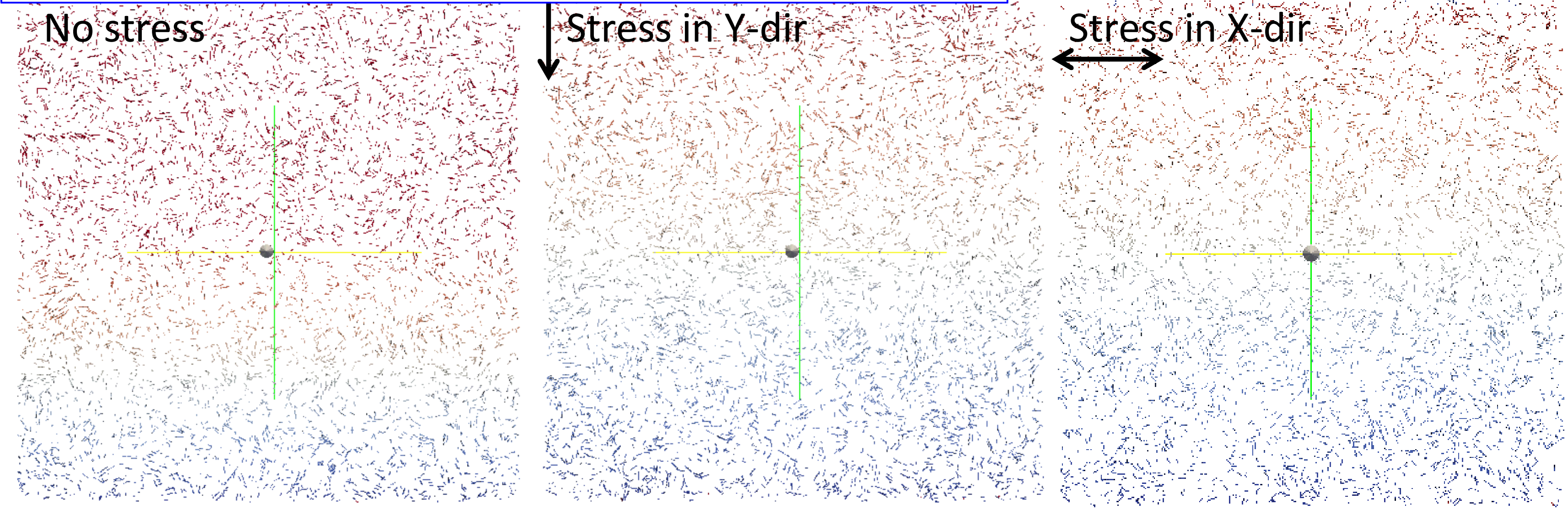
Nucleation and growth of hyd
Simulation resul



Precipitates align themselves perpendicular to the app

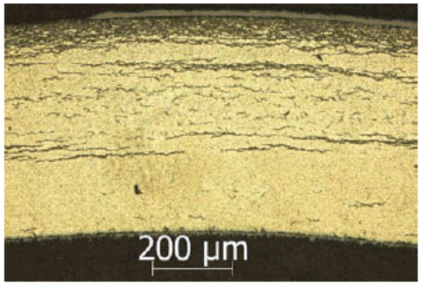
- **No stress, alignment is ~random along basal plane**
- **Stress along Y-dir, alignment is perpendicular**
- **Stress along Z-dir, alignment is *less perpendicular***

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

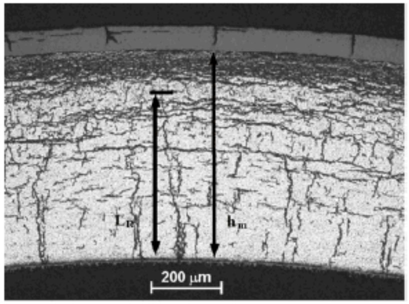


All Zr-based claddings show long-range cooperative formation (Billone et al, ANL UFD report, 2013)

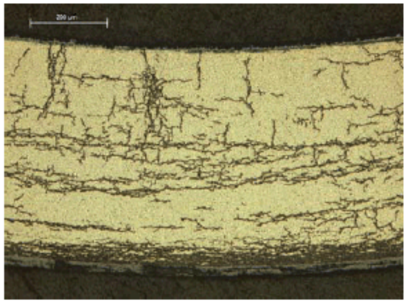
Zirlo™



(a)

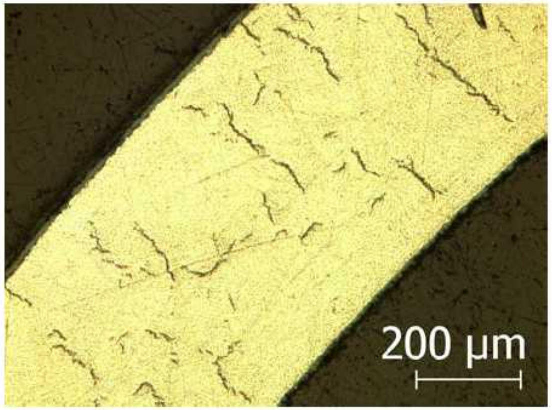


(b)



(c)

M5™



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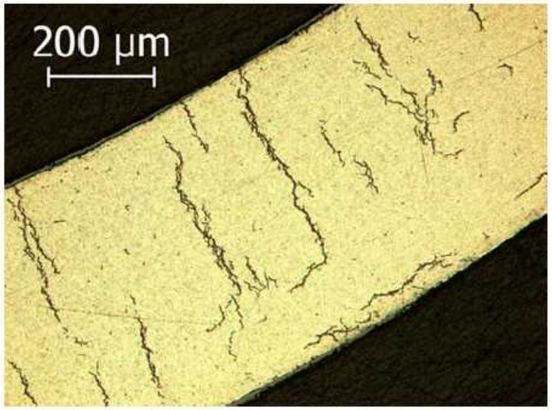
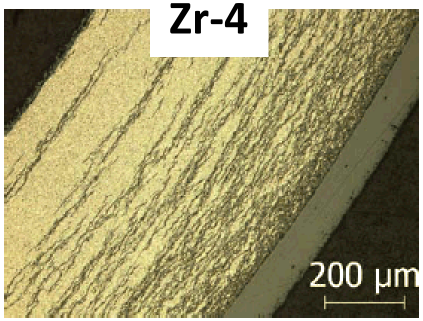
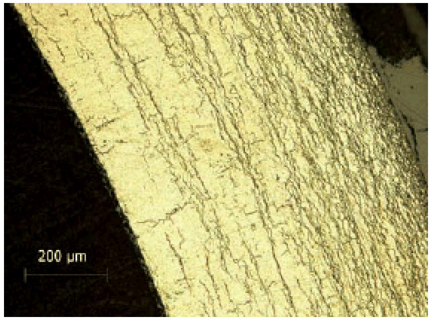


Zr-4



(a)



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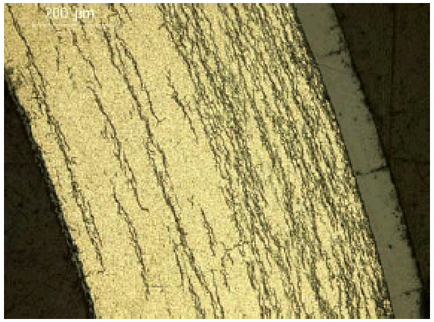

(c)

Conclusions

- **Hydride precipitate re-orientation cannot be simulated by the re-orientation of individual precipitates**
  - **There must be cooperative nucleation and growth of individual precipitates that leads to re-orientation as observed by optical microscopy.**