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# Meso-Scale Modeling of Nuclear Fuel Behavior

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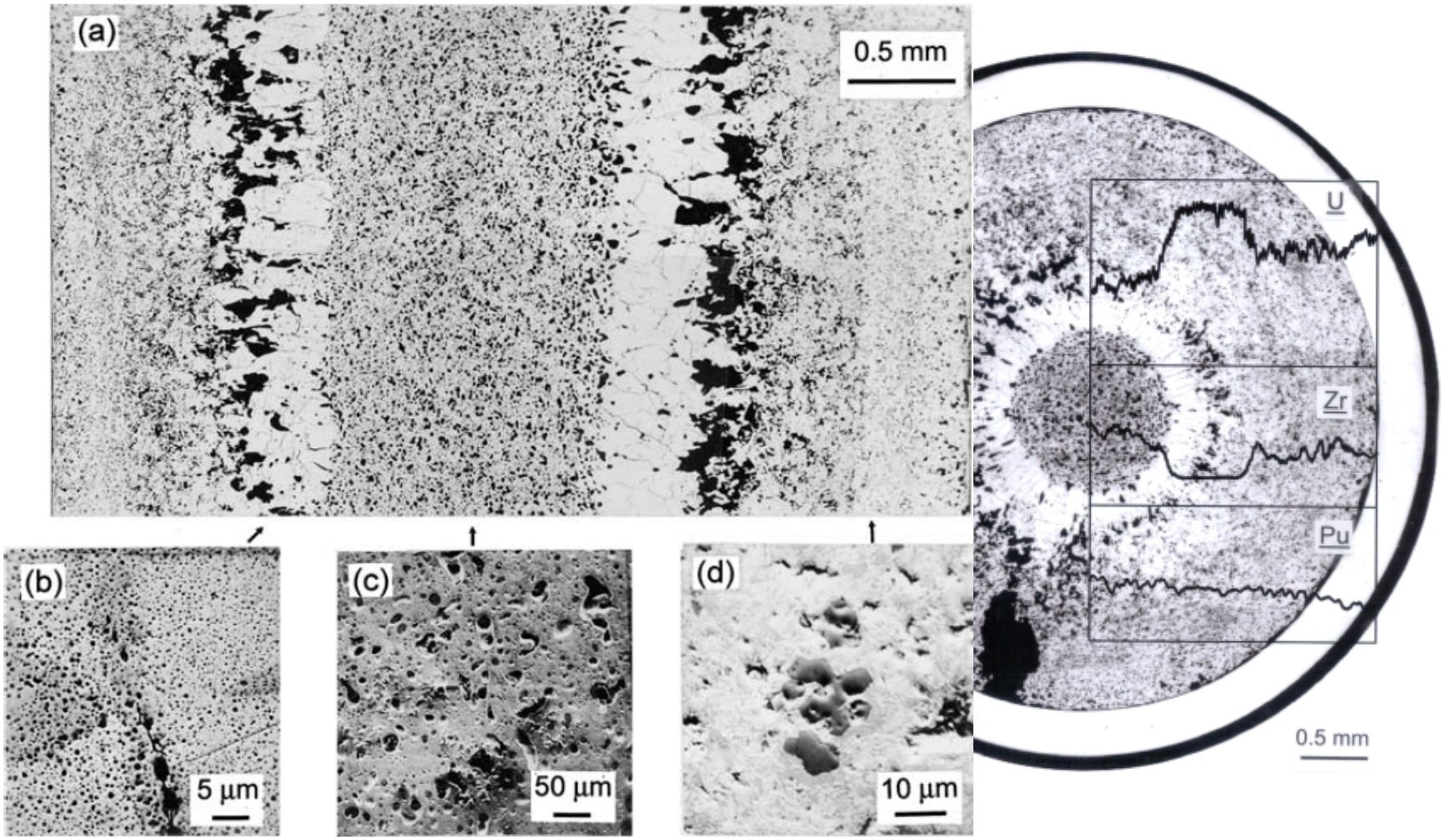
**Advance Nuclear Fuel Cycle Technologies**

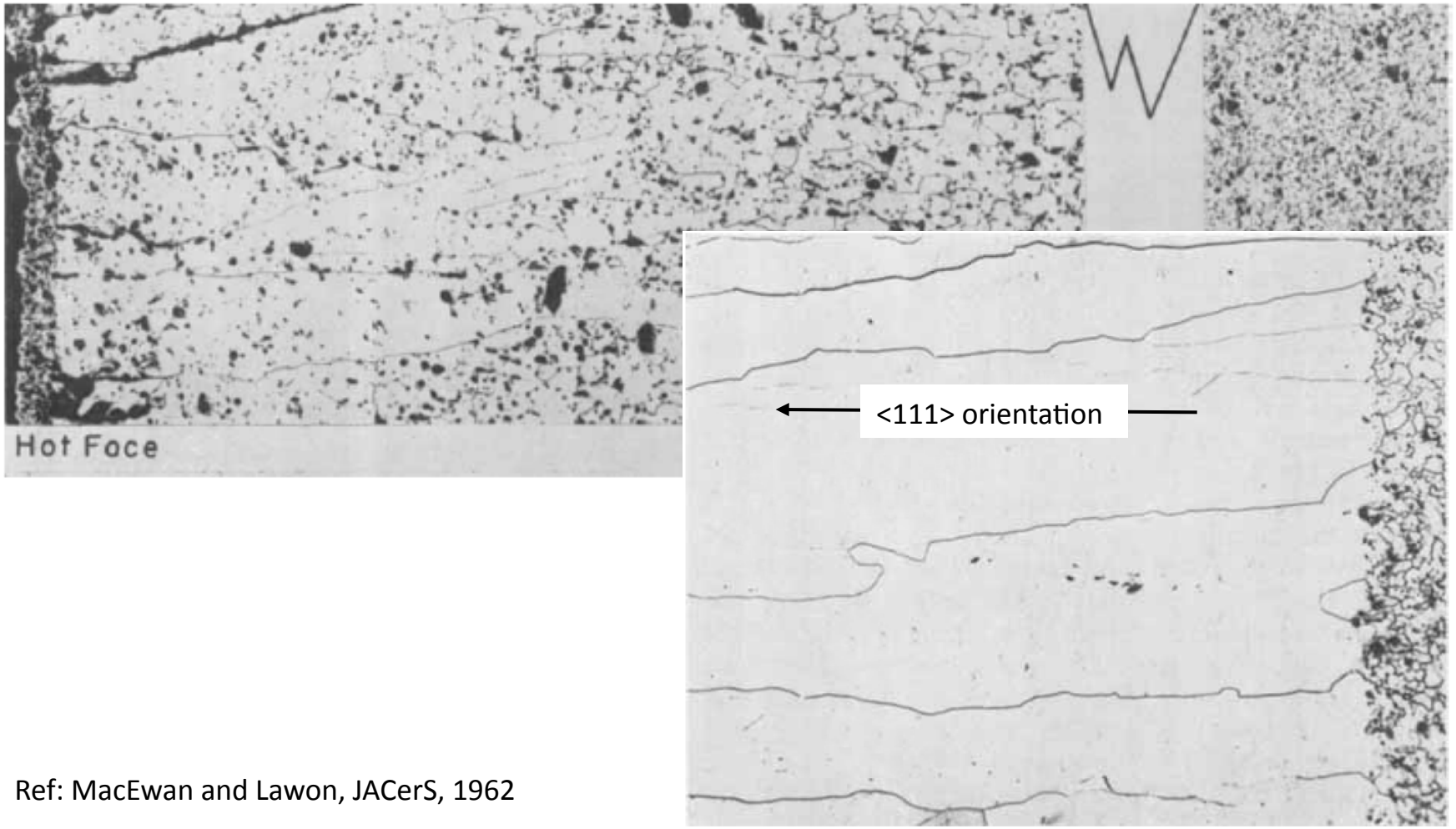
**Albuquerque, NM**

# Segregation, grain restructuring and pore migration in U-Pu-Zr fuel.




Ref: Kim, Hofmann, Hayes & Sohn, JNM, 2004

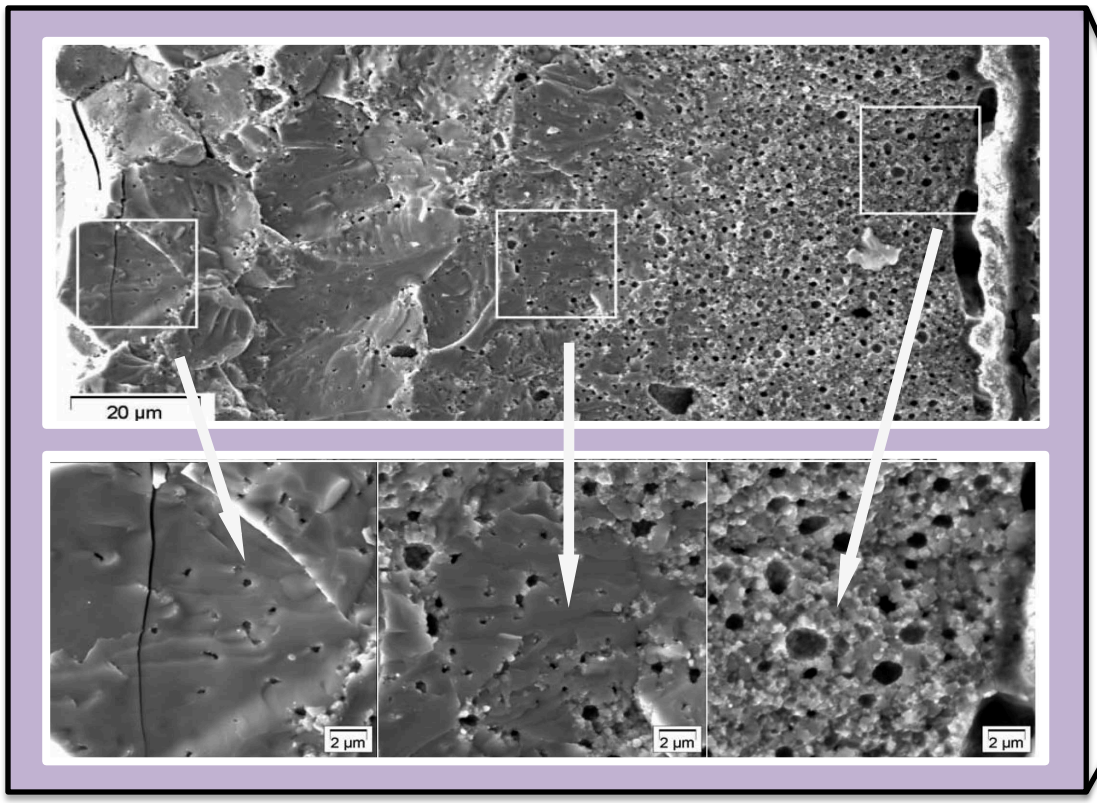
# Out-of-pile experiments showing columnar grain growth and axial pore formation in $UO_2$

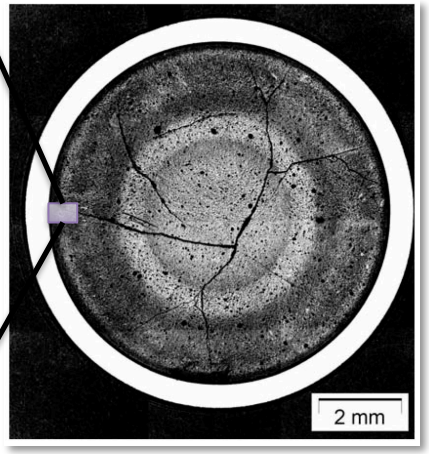



Ref: MacEwan and Lawon, JACerS, 1962

# High-Burnup Rim Structure

- The rim of LWR fuels is enriched relative to the rest of the fuel
- $^{238}U + n \rightarrow {}^{239}U \rightarrow {}^{239}Np + \beta^{-} \rightarrow {}^{239}Pu + \beta^{-}$
- Resulting in more fission events
- Damage accumulates follows fission frequency
- Relatively low temperature causing repeated recrystallization




Irradiated Fuel Cross-Section
*Noirot, et. al* ***NE&T****, vol. 41 2009*

# Although we describe & understand fuel behavior at meso-scale, we model as continuum at macroscale.

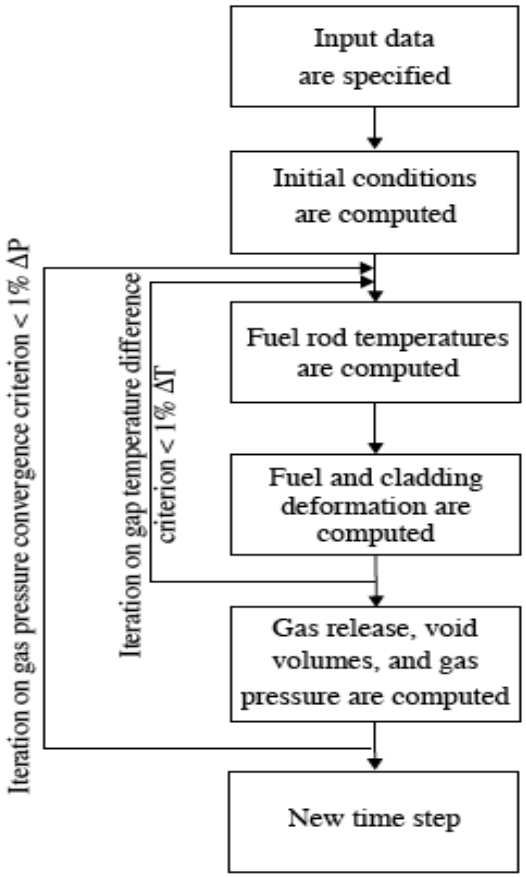



**Figure 2.1** Simplified FRAPCON-3 Flow Chart

## Thermal models in FRAPCON

$$T_b(z) = T_{in} + \int_0^z \left[\frac{4q''(z)}{C_p G D_e}\right] dz$$

Coolant temperature

$$T_w(z) = T_b(z) + \Delta T_f(z) + \Delta T_{cr}(z) + \Delta T_{ox}(z)$$

Clad surface temperature

$$\iint_S k(T,\bar{x})\vec{\nabla}T(\bar{x}) \bullet \hat{n} ds = \iiint_V S(\bar{x}) dV$$

Heat conduction in Fuel pellet

.
.
.

*Ref NUREG/CR-6534, Vol. 2*

# Objective and Outline

## Objective:

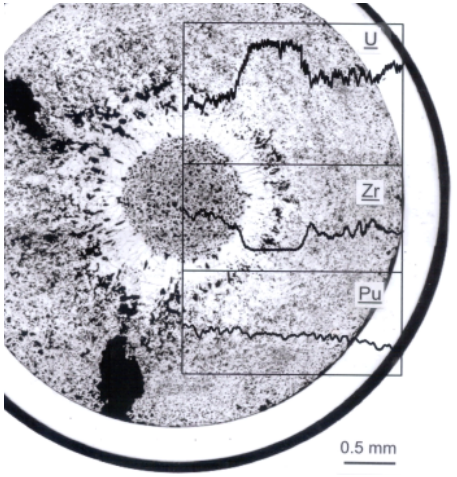
**To demonstrate the utility of meso-scale, microstructural evolution models for studying nuclear fuel behavior.**

## Outline:

- **Review of Potts Monte Carlo and phase field models**
- **Applicability to nuclear fuels**
- **Current trends in mesoscale modeling**
  - **Hybrid models, example is Potts-phase field**
- **Directions of future work**

# Objective: Develop capability for large scale simulation of coupled microstructure-composition problems

- Potts kinetic Monte Carlo, kMC, models are
  - Robust
  - Efficient (very fast)
  - easily incorporate nucleation and phase changes
  - Highly flexible for microstructural evolution
- Phase field, PF, models are
  - Able to incorporate gradient quantities easily
  - Based on thermodynamic functional
  - Highly flexible for compositional evolution problems
- We will develop capabilities that combine the strengths of both for robust simulations of coupled systems.

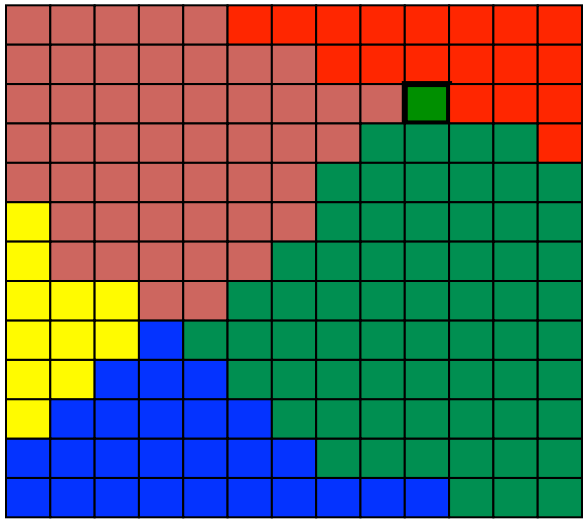



Component segregation*

*Y.S. Kim et al., J. Nucl. Mat., 2004

# 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**



# Equation of State (Thermodynamics)
# Potts kMC

- **Complex EOS can be constructed with two basic components**
  - **A volumetric free energy term, $E_{vol}$**
  - **An interfacial free energy term, $E_{int}$**

$$E = E_{vol} + E_{\text{int}}$$

$$E = \sum_{i=1}^{N}\left( E_{vol}(q_i) + \frac{1}{2}\sum_{j=1}^{n} J\left(1-\delta_{ij}\right)\right)$$

***N*** **– number of sites** ***i, n*** **– number of neighbors** ***j***

  - **These basic components are the basis for grain growth, recrystallization, sintering, abnormal grain growth, etc.**

# Kinetic of Evolution
# Potts kMC

- **Ensemble is statistically manipulated to mimic atomistic diffusive processes.**
- **Boltzmann statistics used for evolution**

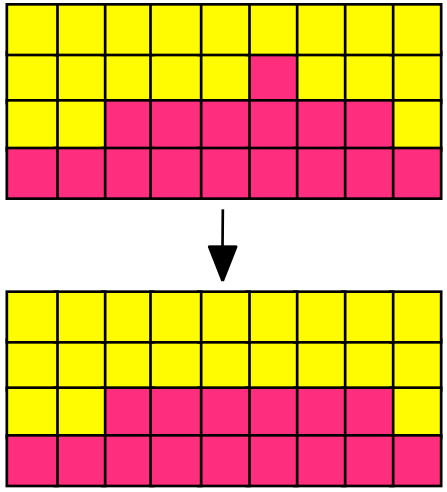
$$P = \begin{cases} 1 & for \quad E \le 0 \\ \exp\left(\dfrac{-\Delta E}{kT}\right) & for \quad E > 0 \end{cases}$$

Evolution of the ensemble

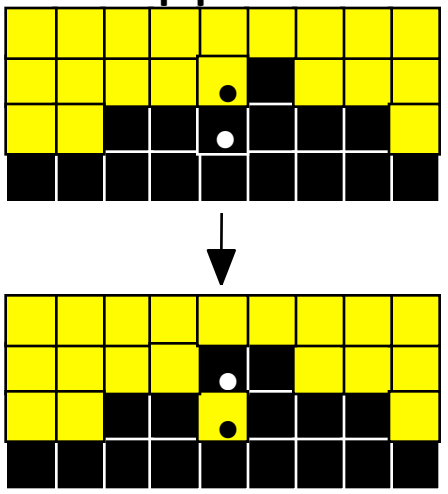
Minimization of Total Free Energy

Kinetics of Microstructural Evolution

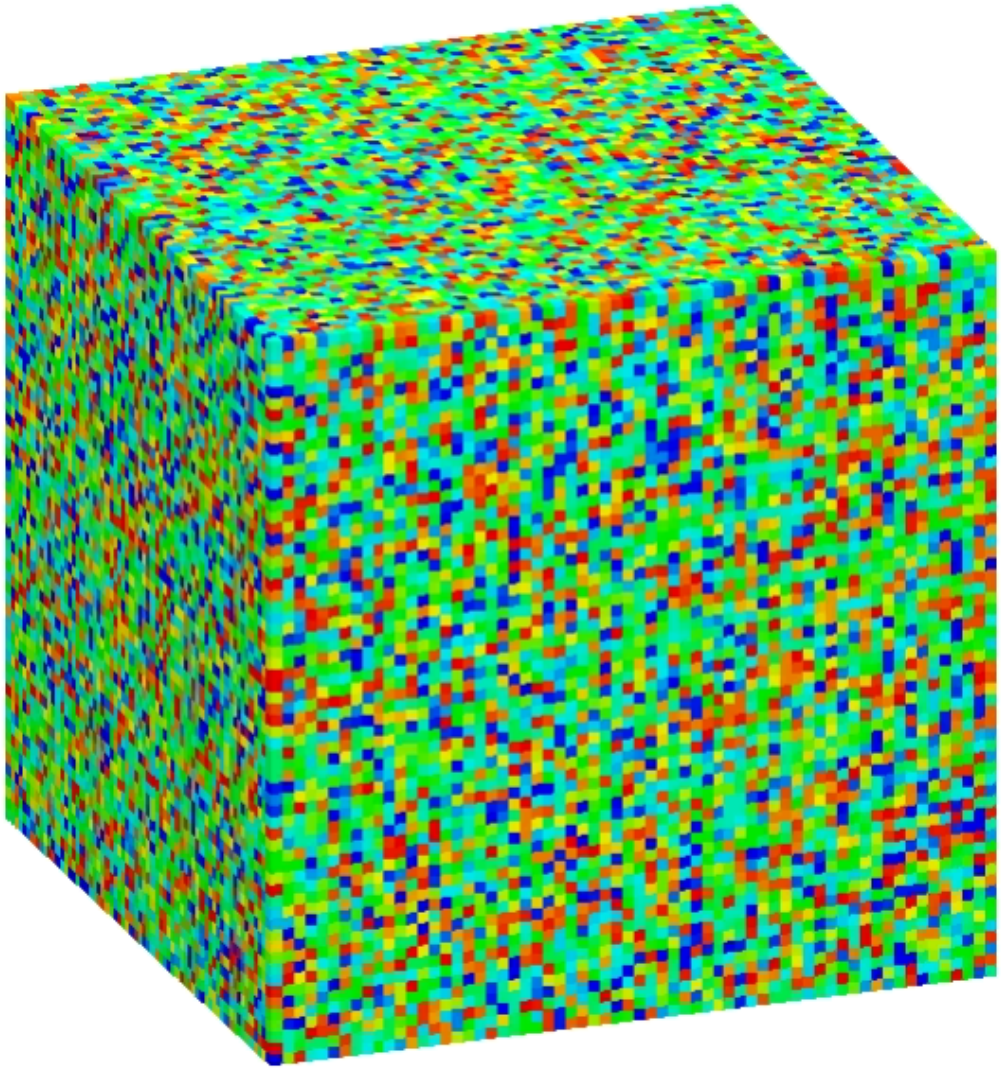
**grain growth**
**change pixel color**



**pore migration**
**vacancy formation**
**swap pixels**



# Potts kMC Simulation of Grain Growth

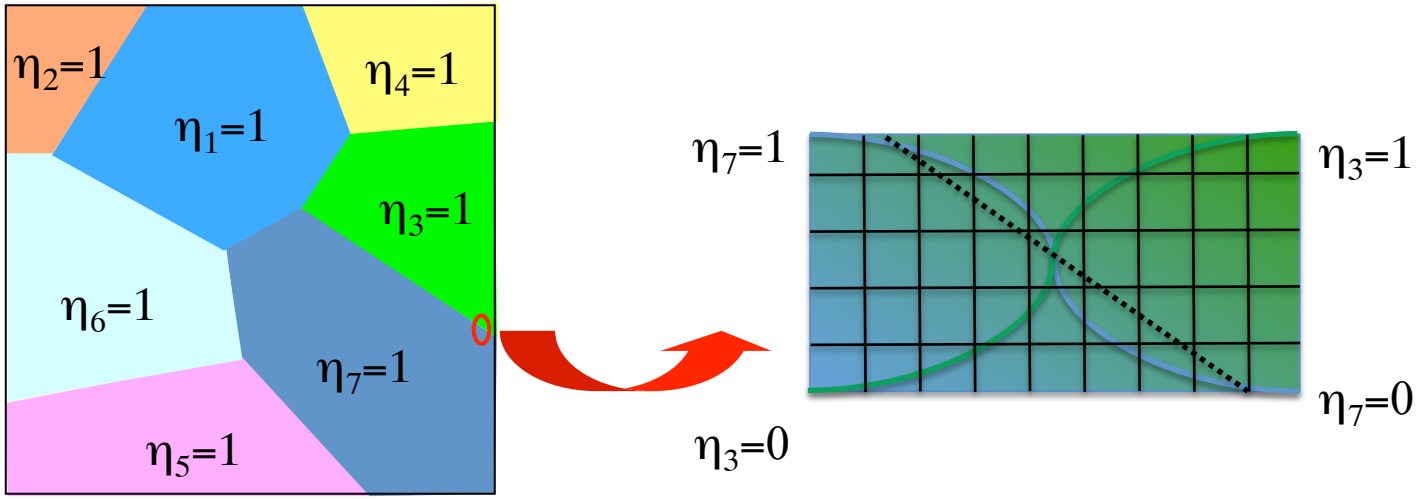


**Shown to simulate**

- **Curvature-driven gg**
- **Correct kinetics, topology and grain size distribution.**

# Representation of Microstructure and Composition Phase Field

- **Phase field maps 'fields' on to a grid**
  - **Each color can represent a membership in a phase, feature and / or composition**
  - **Each lattice point stores the field value, $\eta$ , at that location**
  - **Gradient terms are easy to treat**
  - **Slow and numerically unstable compared to Potts**



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# Equation of State (Thermodynamics)
# Phase Field

- **Again, complex EOS can be constructed with two basic components**
  - **A volumetric free energy term, $f_o$**
  - **An interfacial free energy term, $\sum_{i=1}^{Q} \kappa_{\eta i} (\nabla \eta_i)^2$**

$$E_{pf} = \int \left( f_o + 2 \sum_{i=1}^{Q} \kappa_{\eta i} (\nabla \eta_i)^2 \right) dV$$

  - **These basic components are the basis for grain growth, Ostwald ripening, coarsening of precipitates, etc.**

# Kinetic of Evolution
# Phase Field

$$\frac{\partial c}{\partial t} = -D\nabla^2 \frac{\delta E}{\delta c} = -D\left(\nabla^2\left[\frac{\partial f_o}{\partial C} - \kappa_{\eta i}\nabla^2 C\right]\right)$$

Conserved Eq.

Change Rate

Diffusivity

Driving Force

Mobility

$$\frac{\partial \eta_i}{\partial t} = -M_i \frac{\delta E}{\delta \eta_i} = -M_i\left(\frac{\partial f_o}{\partial \eta_i} - \kappa_{\eta i}\nabla^2 \eta_i\right)$$

Non-Conserved Eq.

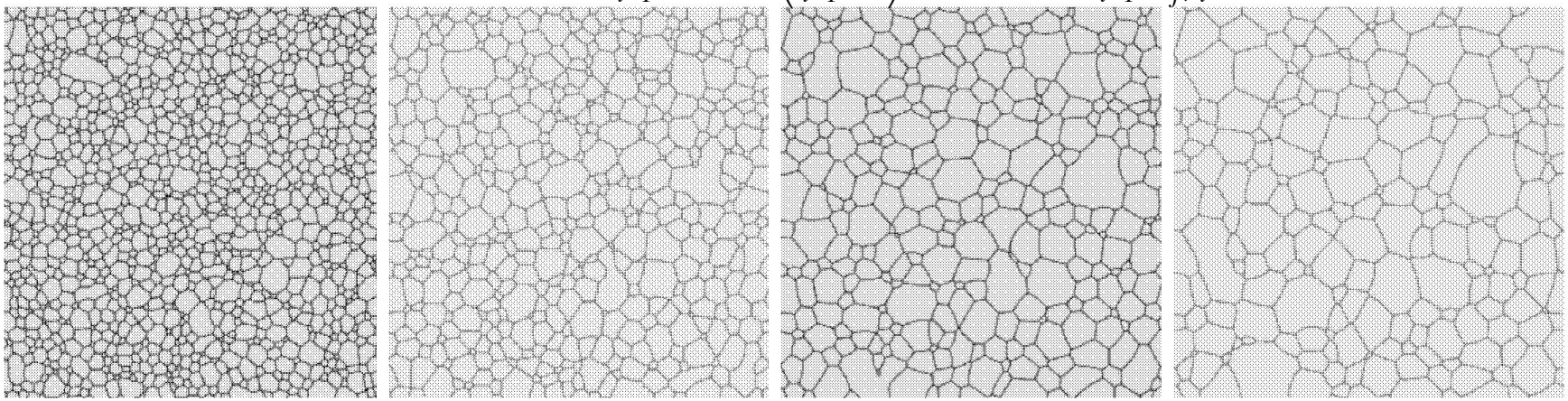
Numerically Solving kinetic equations → Minimization of Total Free Energy → Kinetics of Microstructural Evolution

# Phase Field Simulation of Grain Growth

**Free Energy Functional**

$$F = \int \left[ f_o\left(\eta_1(r),\ldots,\eta_p(r)\right) + \sum_{i=1}^{p} \frac{\kappa_i}{2} \left(\nabla \eta_i(r)\right)^2 \right] d^3 r$$

$$f_o = -\frac{\alpha}{2}\sum_{i=1}^{p}\eta_i^2 + \frac{\beta}{4}\left(\sum_{i=1}^{p}\eta_i^2\right)^2 + \left(\gamma - \frac{\beta}{2}\right)\sum_{i=1}^{p}\sum_{j\neq i}^{p}\eta_i^2\eta_j^2$$



**Phase field model has been shown to simulate**

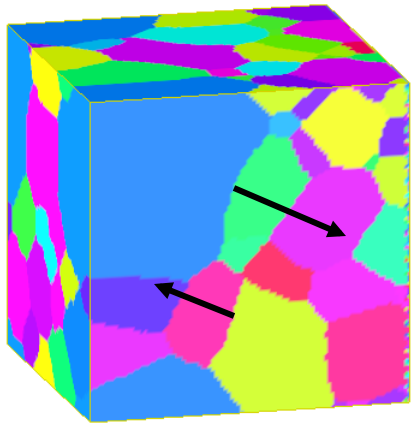
- **Curvature-driven grain growth (with coalescence events)**
- **Correct kinetics, topology and grain size distribution.**

**Computationally intensive**

- **Need ~100 η, so must solve 100 kinetic equations**
- **Need higher resolution, thus grid is finer.**

# Grain Growth in a Thermal Gradient

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



$$v = -M\kappa$$

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

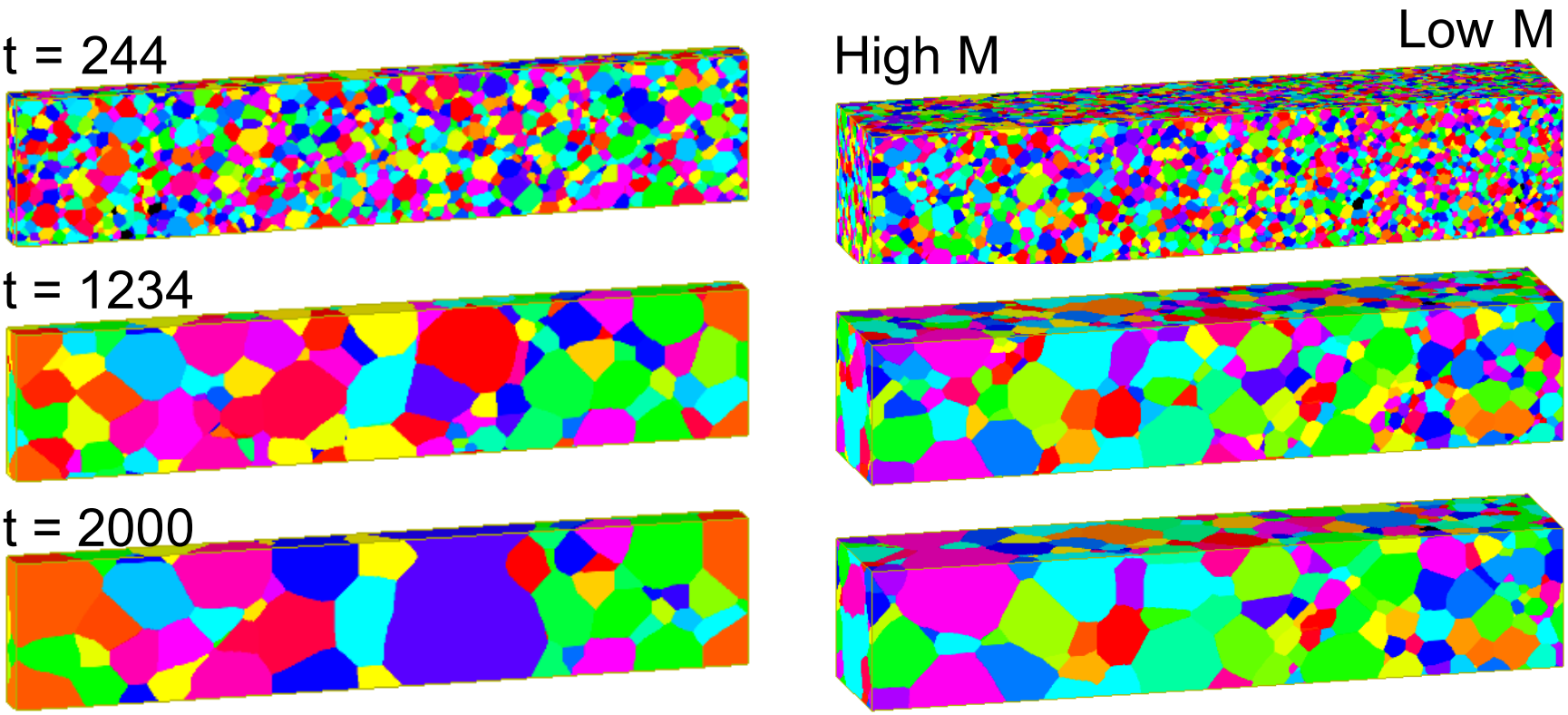
**Thermal gradient is implemented in the model by making the probability of grain growth events a function of temperature**

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

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

# Polycrystalline grain growth in a thermal gradient

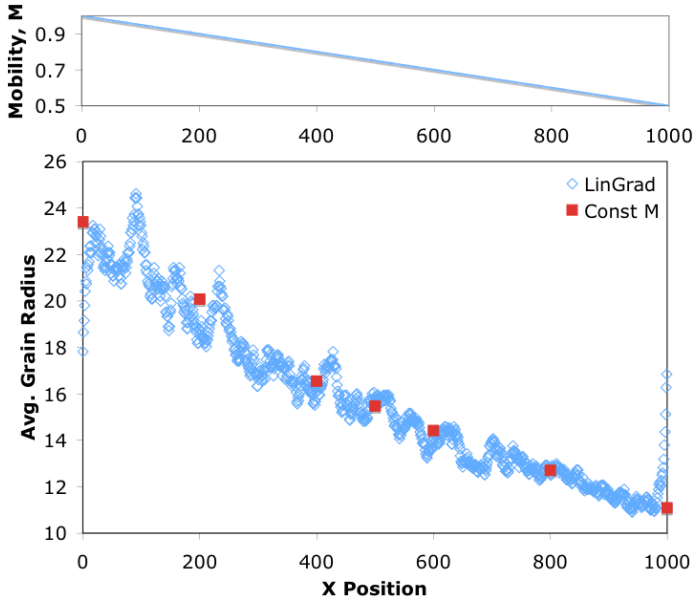
**Isothermal grain growth in compared to one with linear mobility gradient**

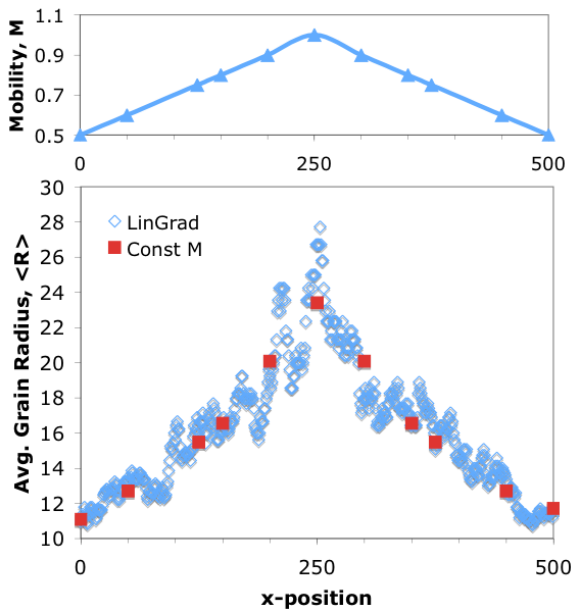



**As expected, grains grow faster in the high temperature region where grain boundary mobility is high.**

# 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.**

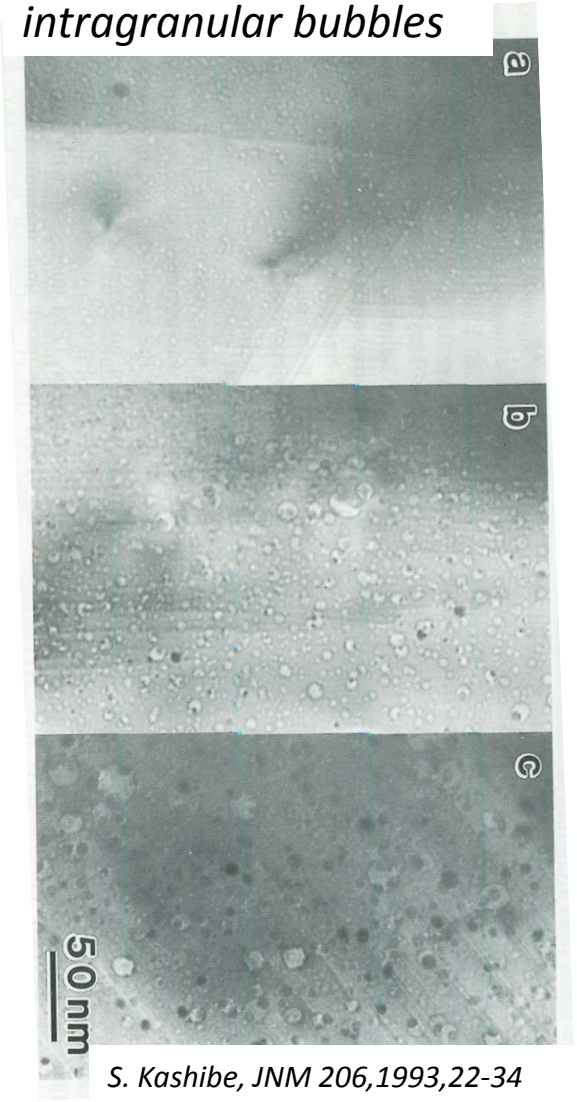



**In smoothly varying thermal gradients, the kinetics of grain growth are locally normal.**

# Gaseous Fission Products Accumulation & Release

*intragranular bubbles*

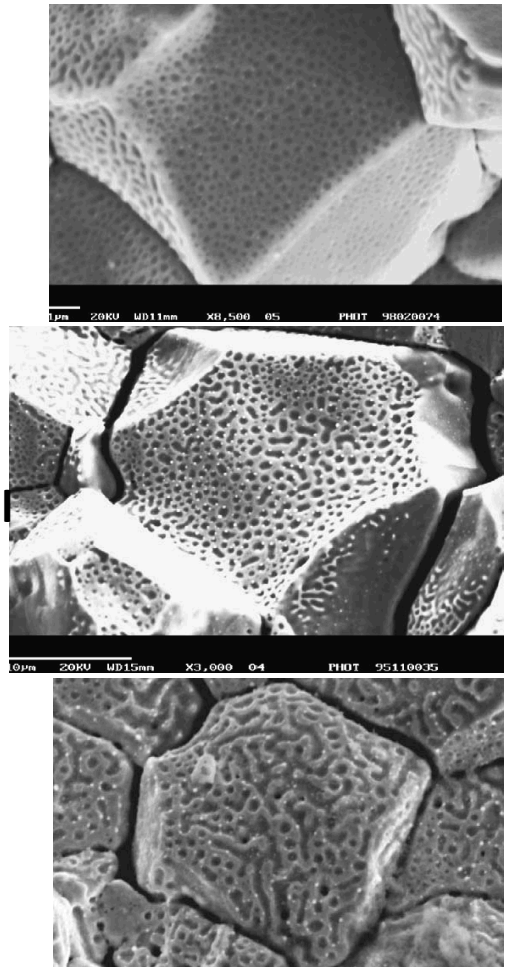



*S. Kashibe, JNM 206,1993,22-34*

**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*



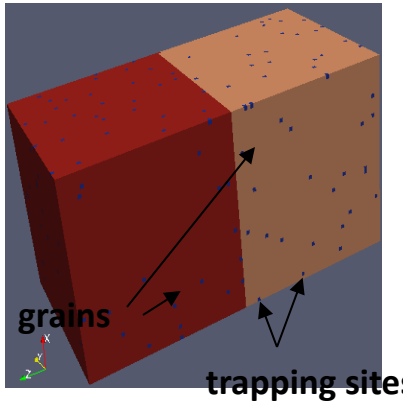
*R.J. White, JNM 325,2004,61–77*

# The model simulates fission gas generation and transport

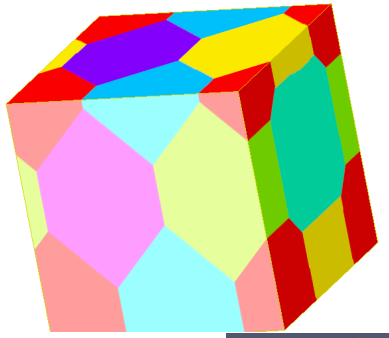
| Reality | Model |
| --- | --- |
| • Fission gases are generated in proportion to fission events<br>• They diffuse in the fuel lattice until precipitating in bubbles<br>• Two sets of bubbles: inter- and intra-granular bubbles<br>• They precipitate on trapping sites (defects due to irradiation) to form nano-sized intragranular bubbles<br>• They precipitate on grain boundaries to form grain boundary bubbles<br>• Fission fragments and neutron irradiation re-dissolve the gases from the bubbles. | • Gas atoms are generated in proportion to fission events<br>• They diffuse by random walk with D(T, damage, C, …)<br>• Two sets of bubbles: inter- and intra-granular bubbles<br>• They precipitate on trapping sites (stationary nucleation points) and diffuse along the surface<br>• They precipitate on grain boundaries and diffuse and coalesce on the gb to form bubbles<br>• Fission fragments and neutron irradiation re-dissolve the gases from the bubbles. |

## Model developed in stages by applying to increasingly more complex geometries

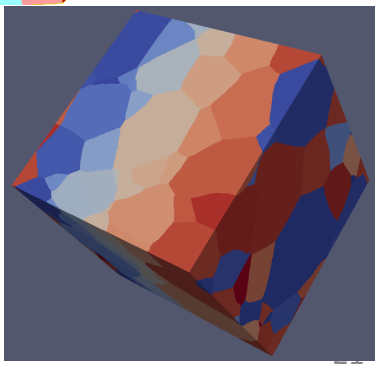
1. Two grain geometry with only grain boundary faces
   - Used to develop simulation parameters
   - Study percolation on planar grain boundary with realistic bubble topology




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

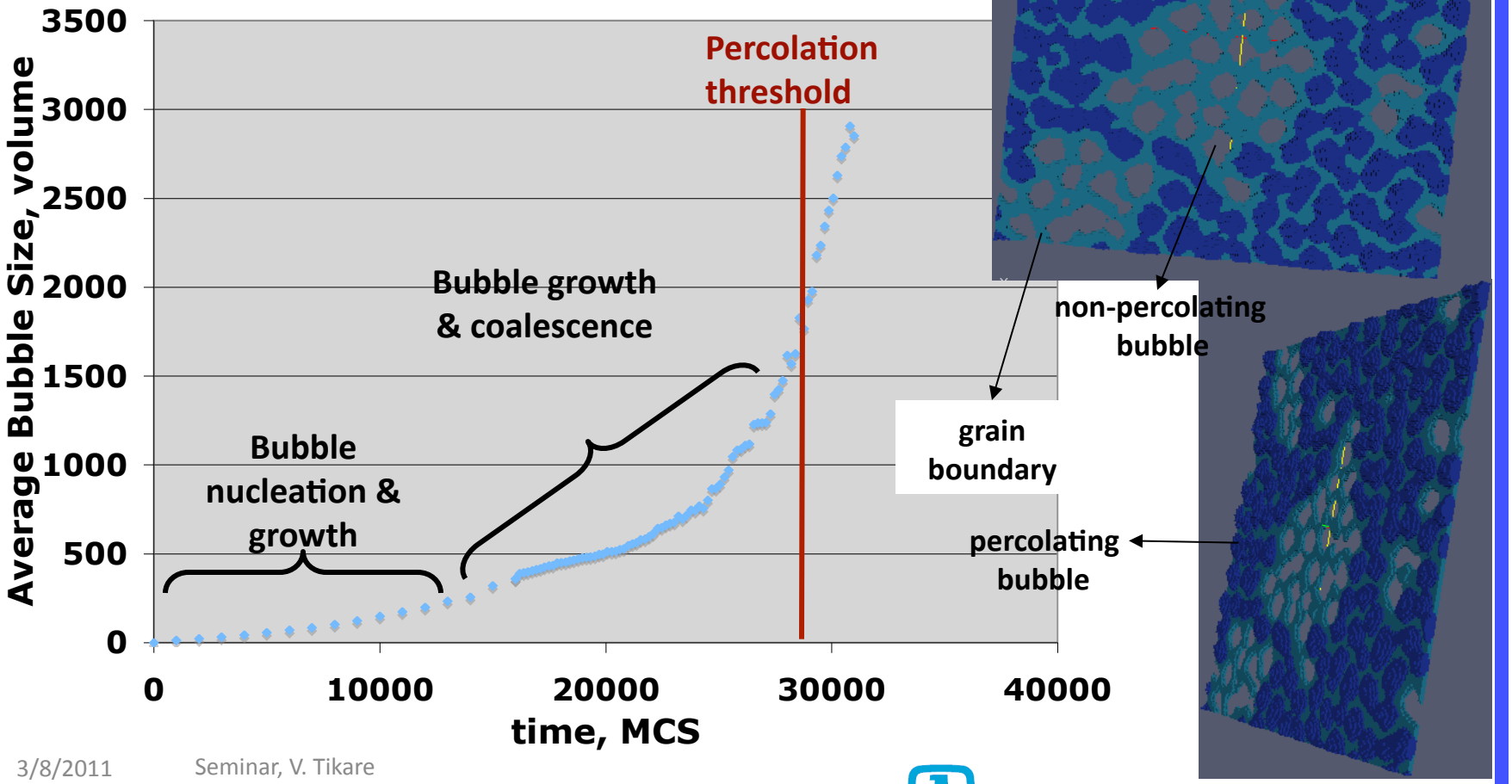


3. Topologically correct polycrystalline geometry
   - Introduce all the complexity of polycrystalline grains
   - Faces of different size, shapes curvatures,
   - edges of different lengths and curvatures
   - corners at varying distances



# Bubble growth at semi-infinity grain boundary

**Regions of bubble growth can be identified:**



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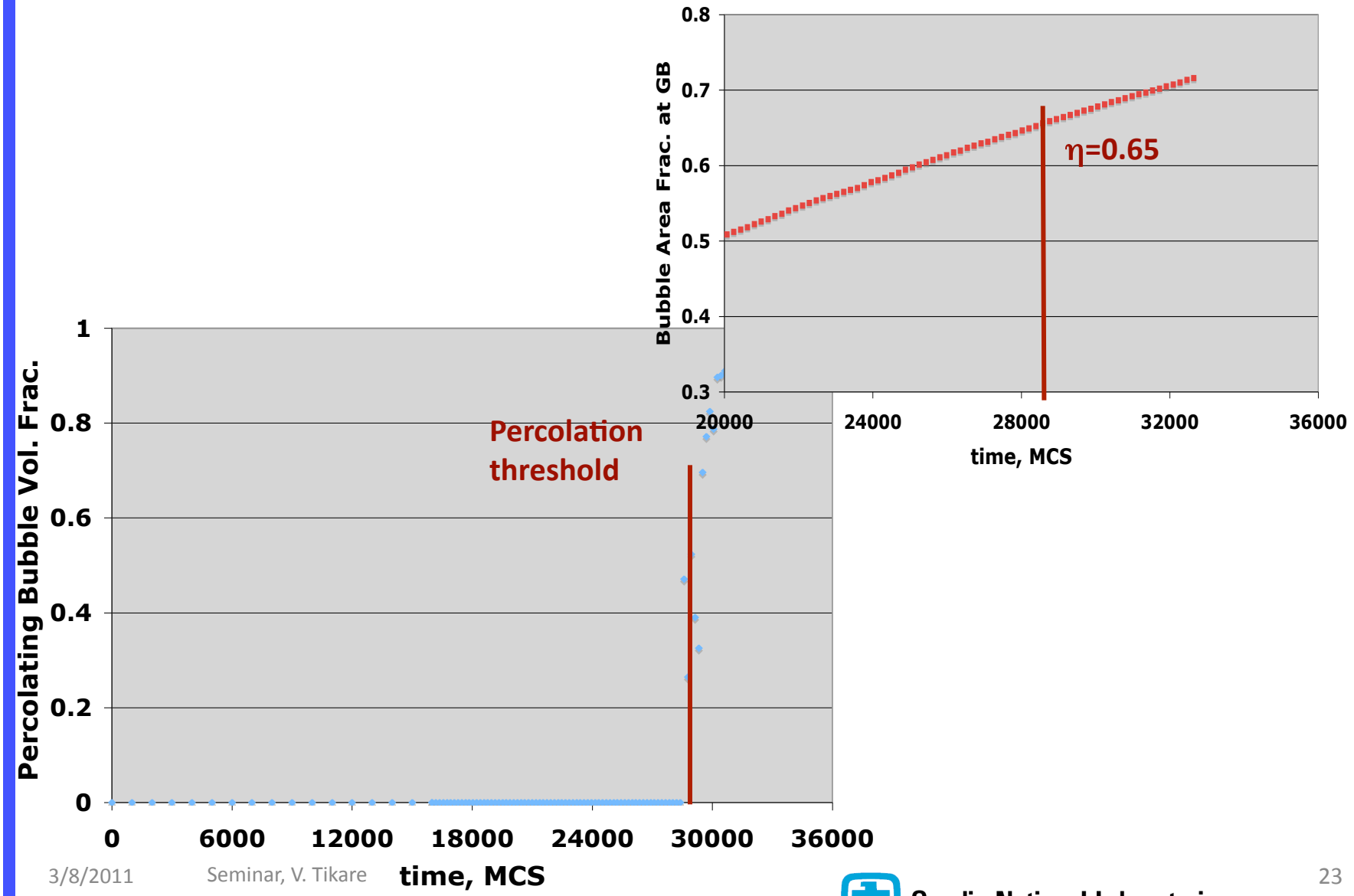
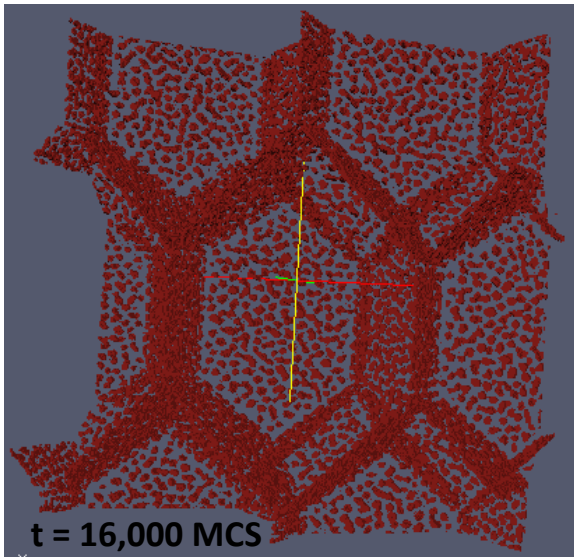
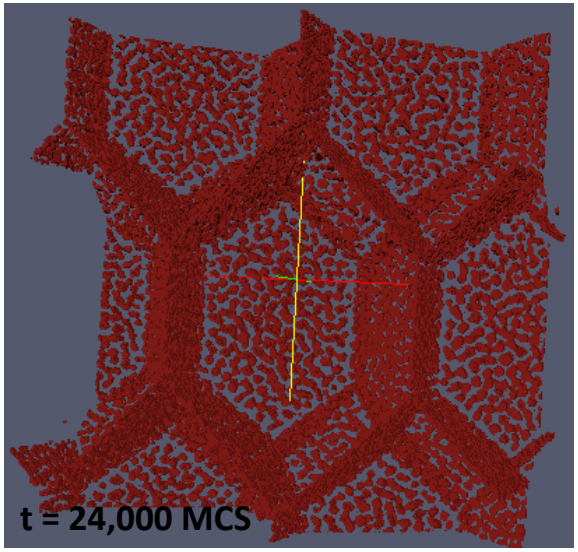
# Bubble growth at grain boundary



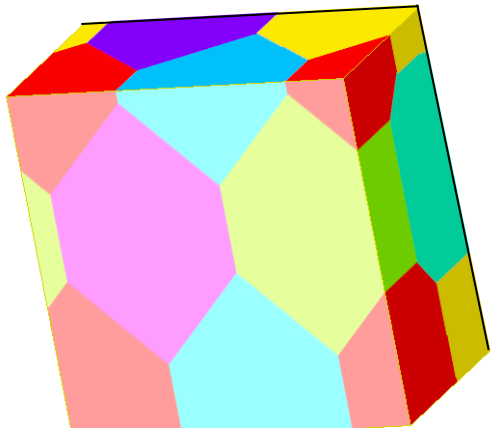
# Fission Gas Bubble Behavior in CP TK Grains

Bubbles at GBs



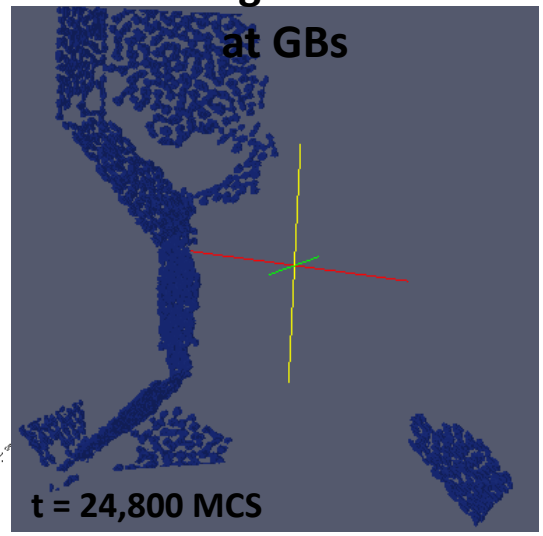

Half the cube is imaged

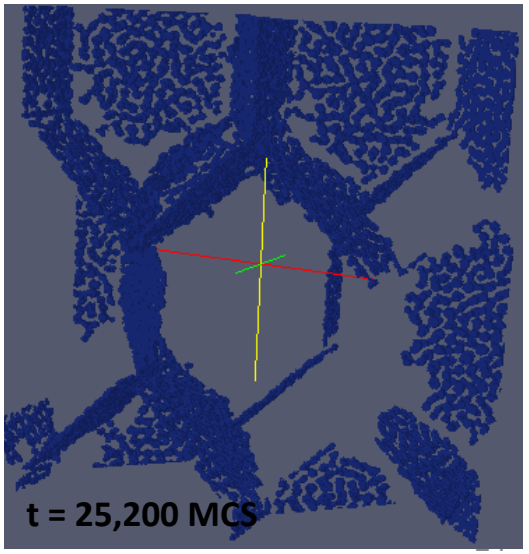


The same 3 regions
- nucleation & growth
- growth & coalescence
- percolation

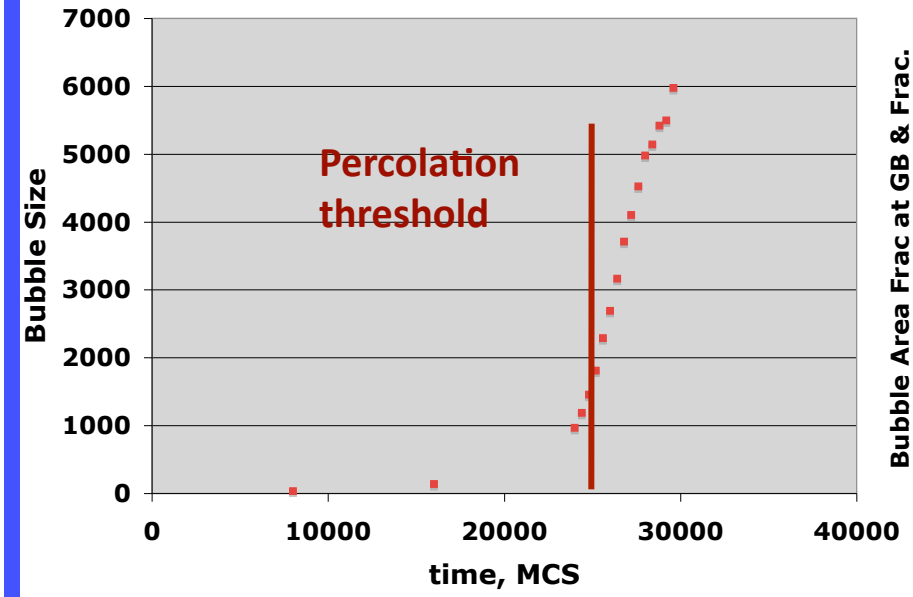
are observed in poly-crystalline fuel grain boundaries.

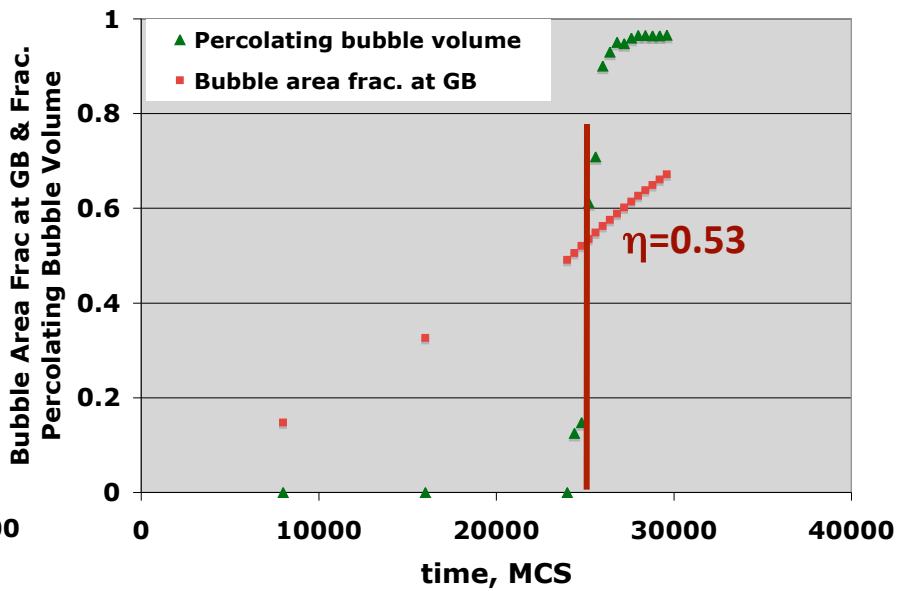
Percolating bubbles at GBs



# Fission Gas Bubble Behavior in Tetrakaidecahedral Grains




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

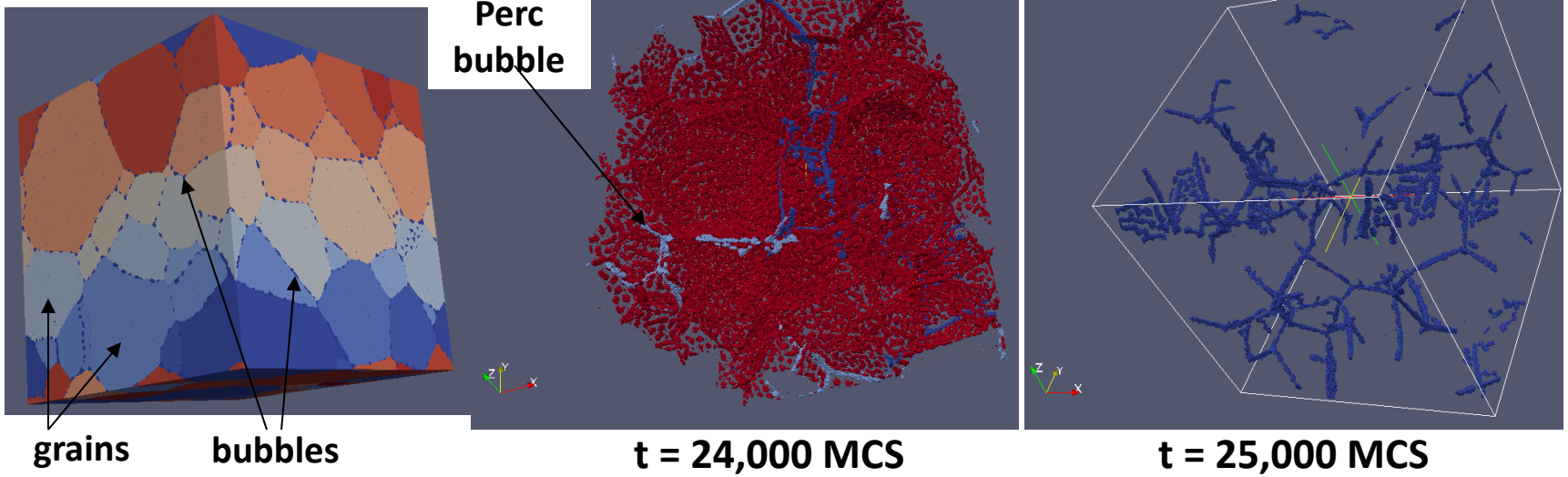
- **3 regions of intergranular bubble growth**
- **Percolated at *lower gb coverage* $\eta$=0.53**
- **Grain edges lower percolation threshold**

# Fission Gas Bubble Behavior in Polycrystalline Grained-Fuels

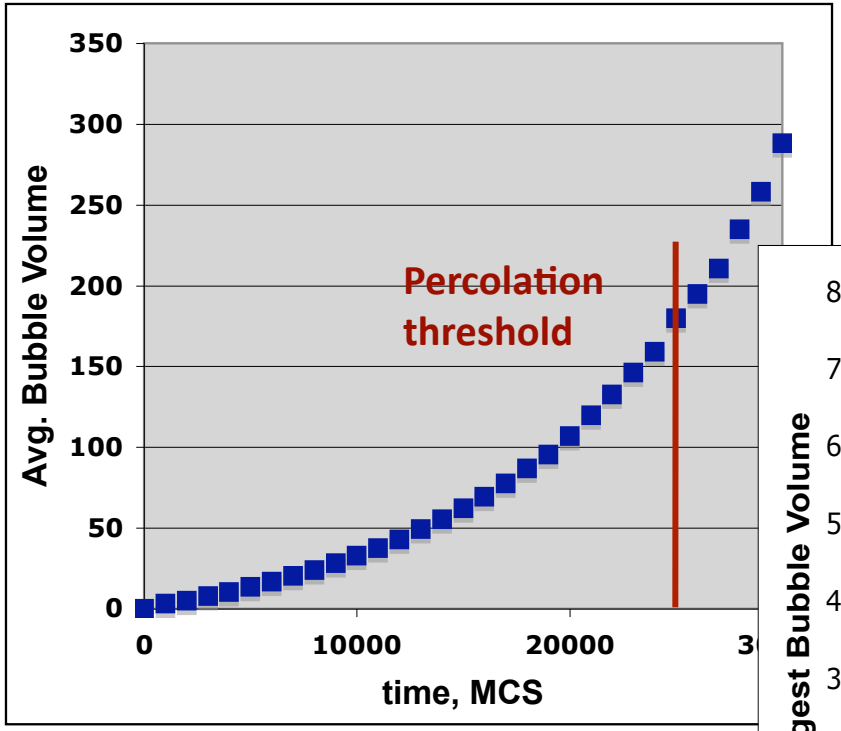
**Bubble growth and percolation in a polycrystalline fuel**

- with distribution in face and edge size and shape.

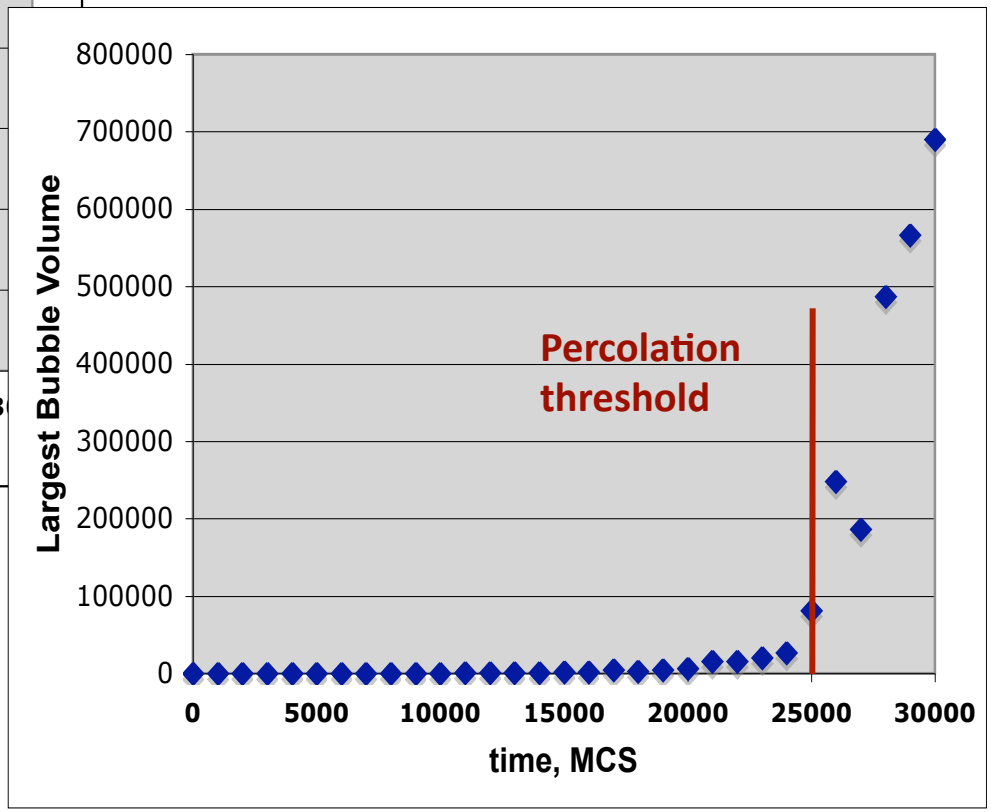
- **Bubble shape at edge is ellipsoidal**
- **Bubble shape at corners is tetrahedral**
- **Linkage among edge & corner bubbles occurs before that on grain boundary faces**



# Fission Gas Bubble Behavior in Polycrystalline Grained Fuels



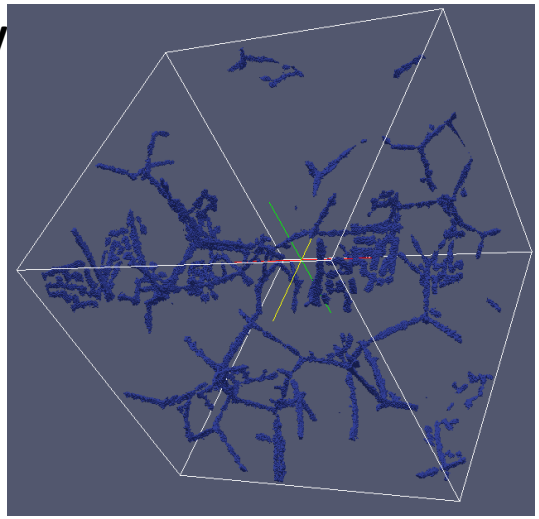

Grain boundary coverage at percolation is $\eta = 0.47$




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

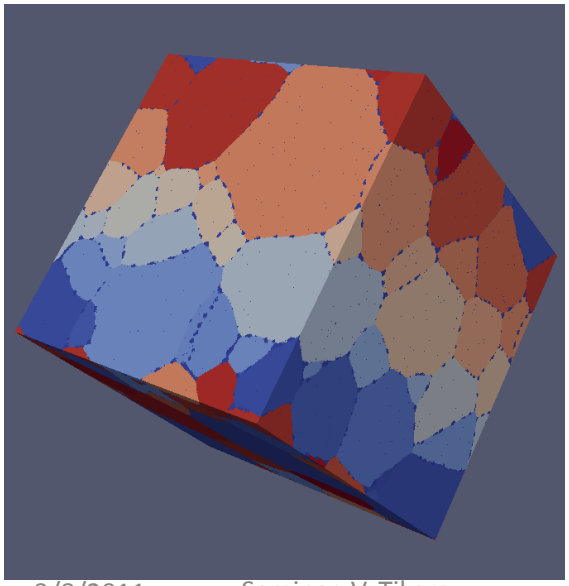
When percolation occurs, the polycrystalline has a large inventory of gas in gb bubbles.

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

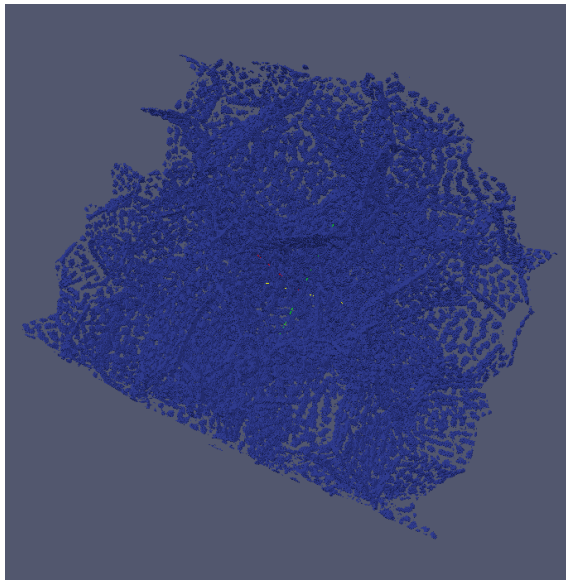
Gas generation and transport continue.

This bubble is sintered but the rest are left.

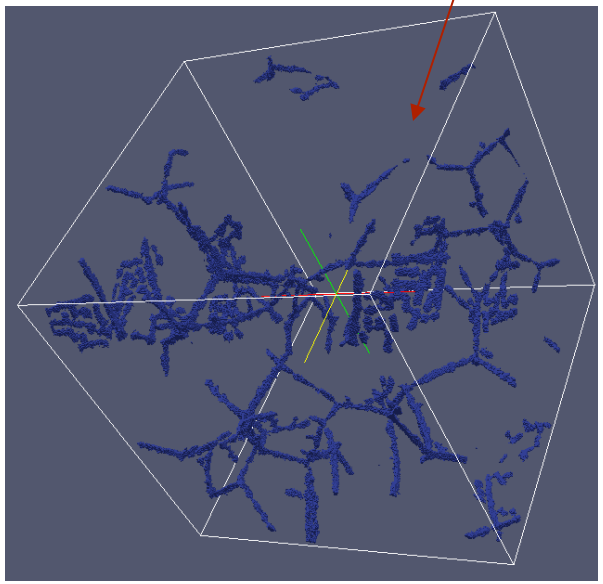
Gas bubbles in polycrystal



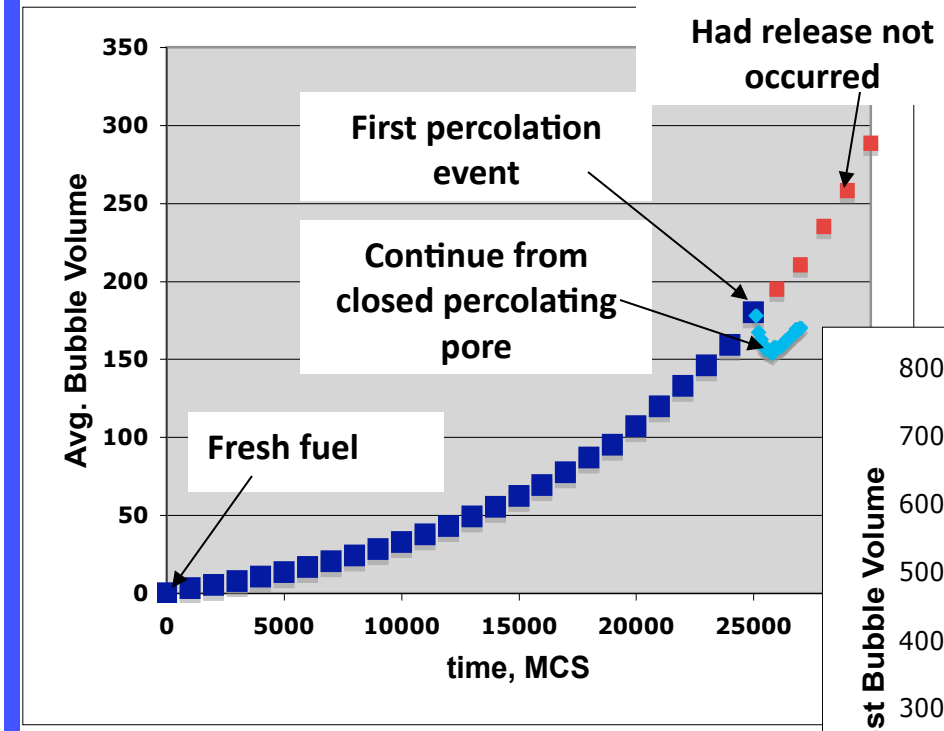
Gas bubbles only



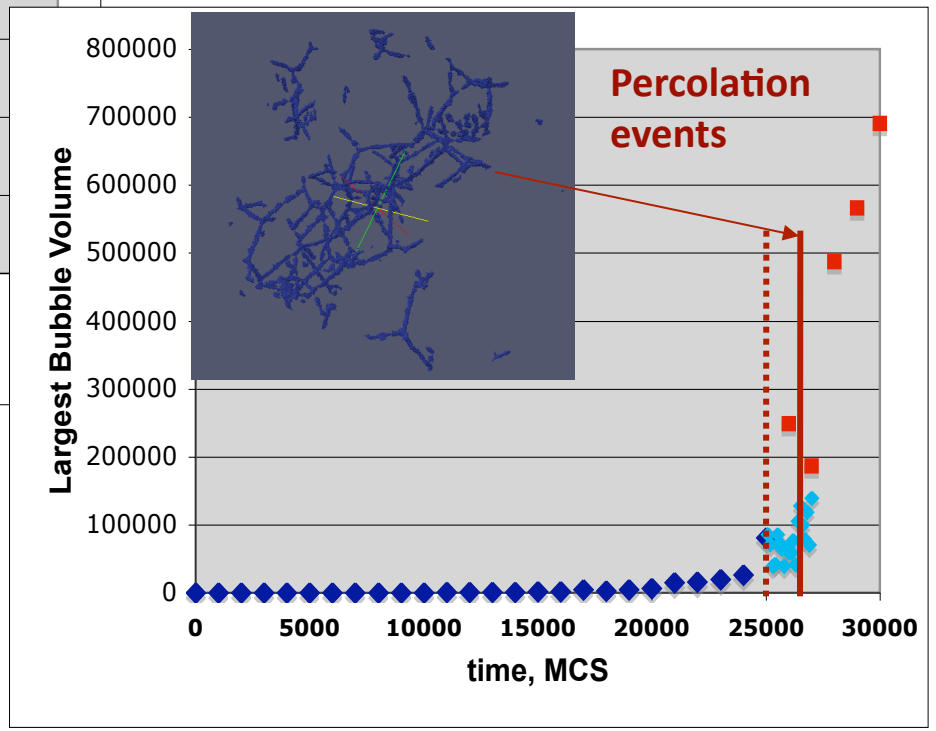
Percolating gas bubble only



# 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 $\psi = 0.096$ at gb coverage $\eta = 0.44$




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



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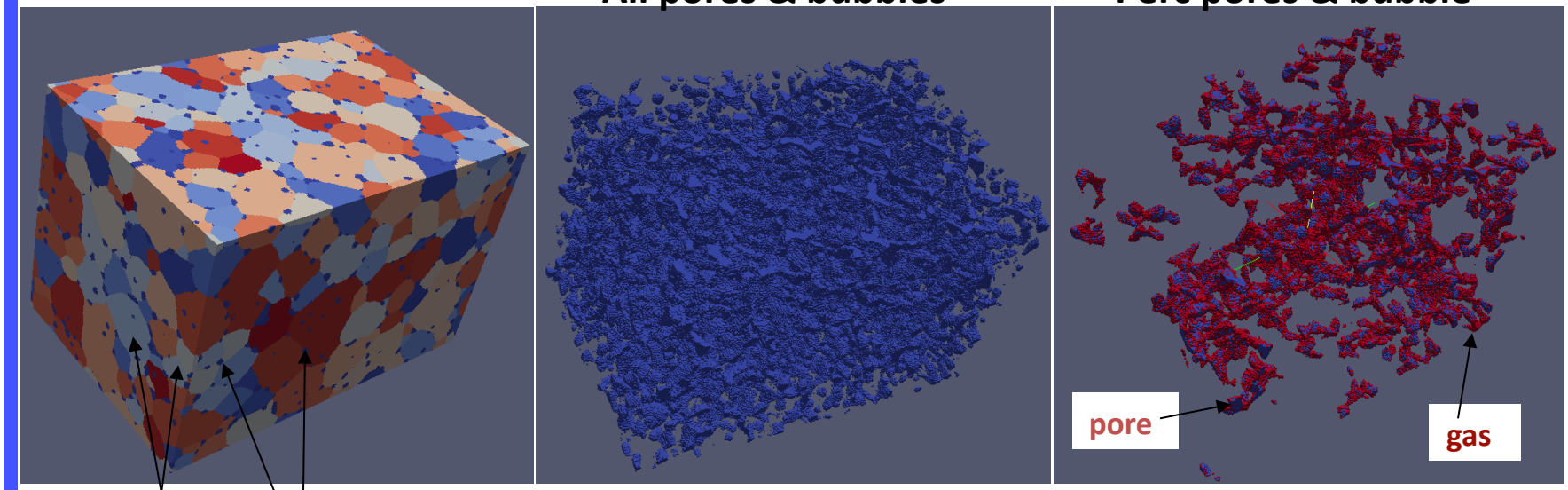
# Fission gas formation and transport in porous polycrystalline fuel

**Preliminary results for polycrystalline with 5% porosity**

- **Pores are intergranular, mostly at quadrajunctions**
- **Bubble form and some link with pores**
- **Pores provide a sink for gas**
- **Percolation still primarily along edges**

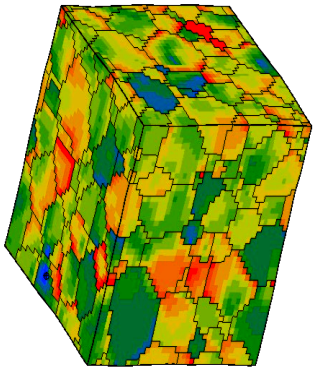
**All pores & bubbles**

**Perc pores & bubble**

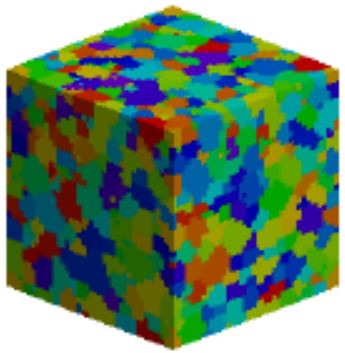



**grains** **pores**

# Potts kMC materials microstructure evolution modeling is a highly developed technique

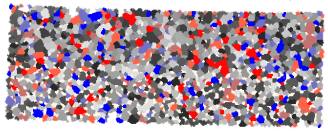


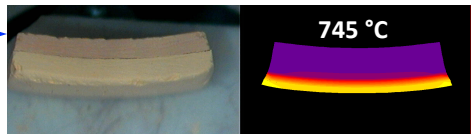
Crystal Plasticity



Ceramic Sintering

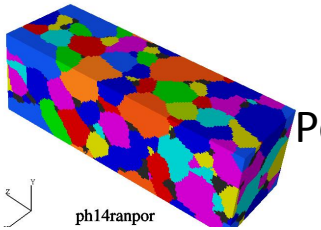
Bi-layer Microstructure



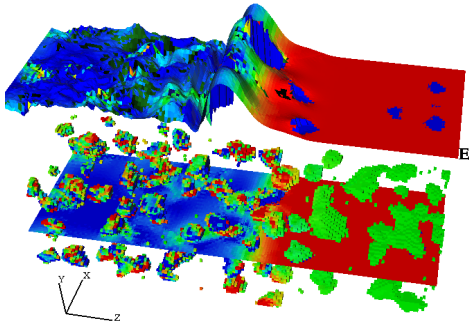



Experiment FEA

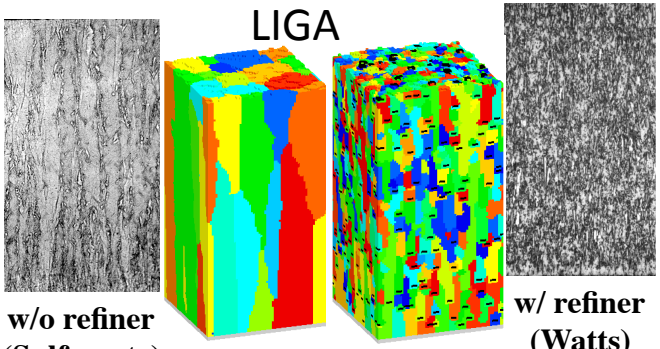
**Microstructure**




Shock Response in Porous Ferroelectrics



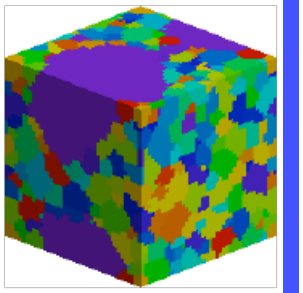
Electrodeposition / LIGA

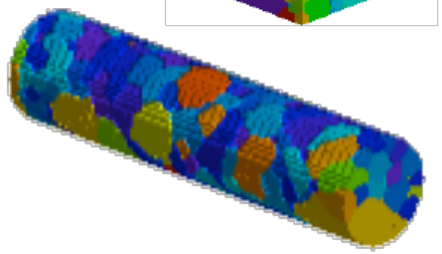


w/o refiner (Sulfamate)

w/ refiner (Watts)

Abnormal Grain Growth & Recrystallization

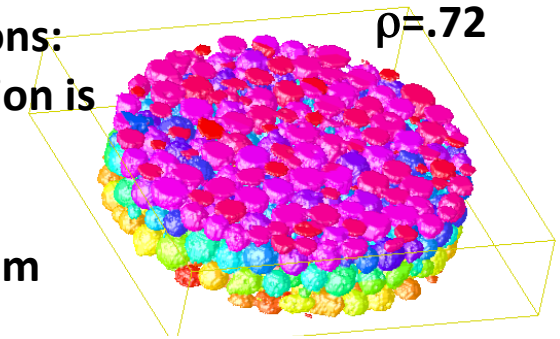


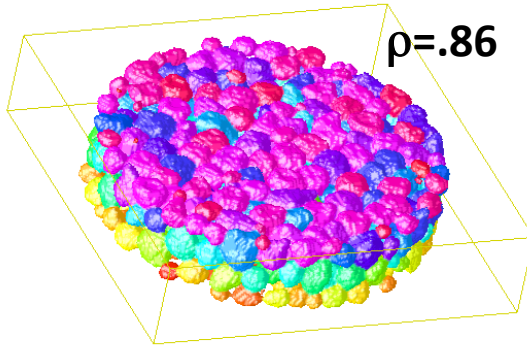


# Current Trend in Potts Materials Modeling: Hybrid Models

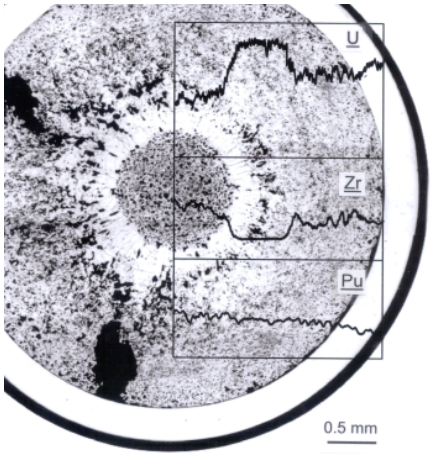
**While Potts kMC is highly developed, it does have limitations:**

**•It is an energy minimization model, thus plastic deformation is difficult to handle. Sintering of ceramics, swelling of fuels.**



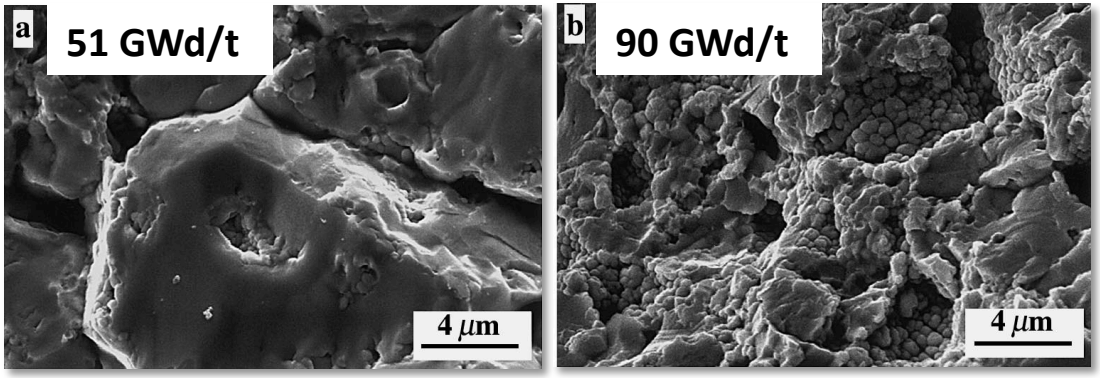

**•Limited diffusion ability as diffusion is simulated by random walk of individual sites**




**•Some coupled processes such as recrystallization which involves energy dissipation give incorrect kinetic results.**

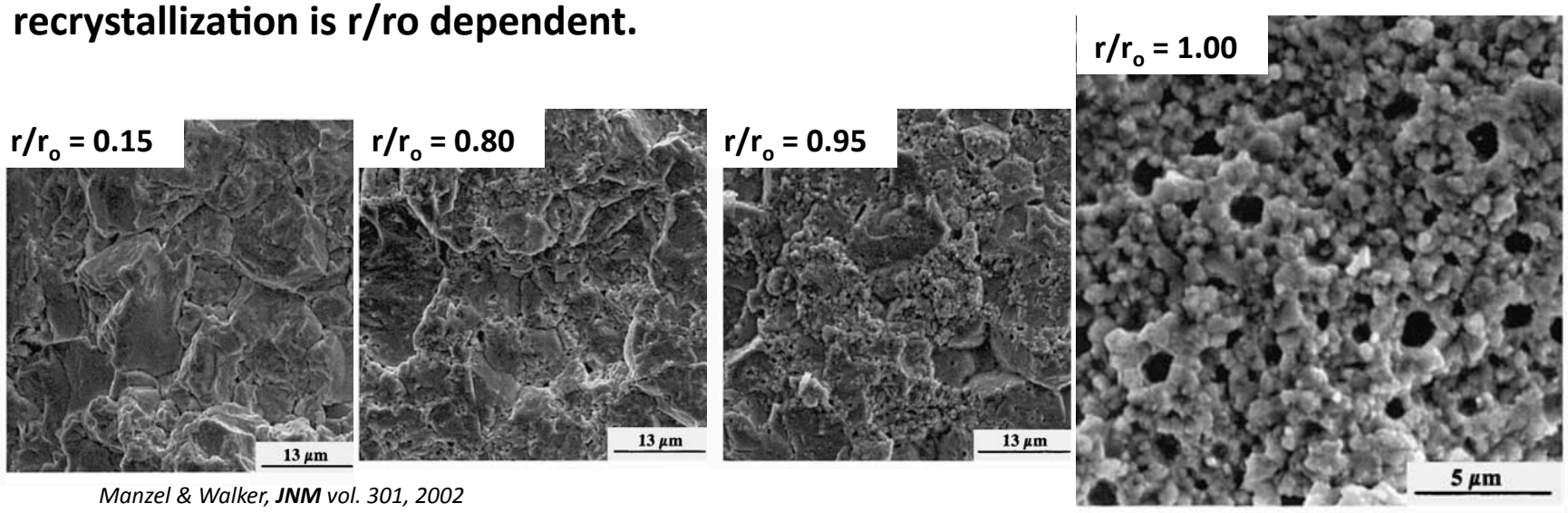
# High Burn-Up Structure

- Rim structure is a function of burn-up, temperature
- At low T, recrystallization becomes the energy dissipation mechanism




*Une, et. al, **JNM** vol. 288, 2001*

- Enrichment, burn-up & temperature are radially dependent thus recrystallization is r/ro dependent.




*Manzel & Walker, **JNM** vol. 301, 2002*



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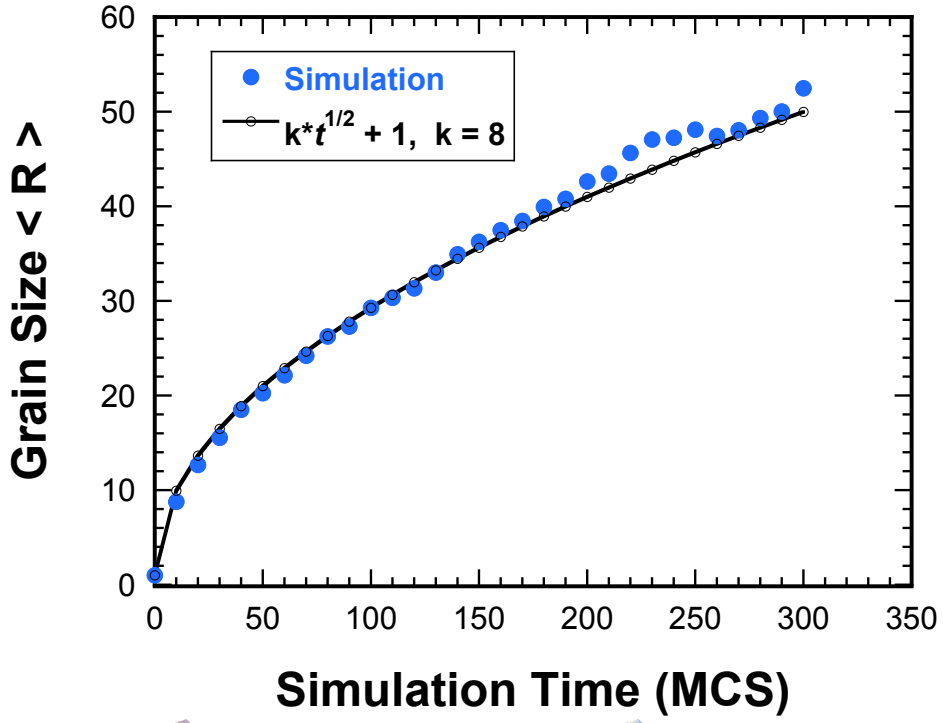
# Recrystallization Model: Hybrid Potts-Cellular Automata

- Potts kMC does not simulate recrystallization with correct kinetics (JMAK kinetics)

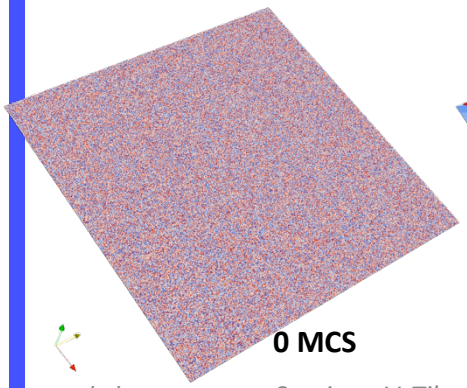
$$f = 1 - \exp\left(-Kt^n\right) \qquad K = -\frac{\pi}{3}\dot{N}G^3$$

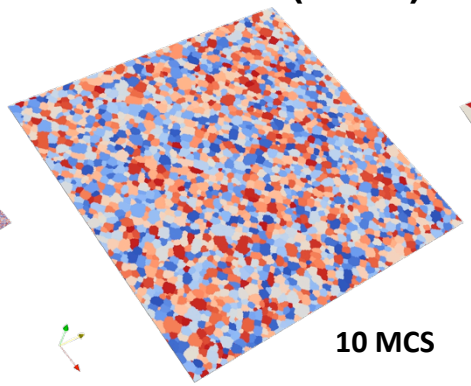
- Raabe & Rollett introduced a hybrid Potts-Cellular model
  - Curvature-driven grain growth by the Potts model
    - Grain growth driven by interfacial energy
  - Recrystallization by CA
    - Nucleation determined by a rule
    - Grain growth by bulk energy minimization

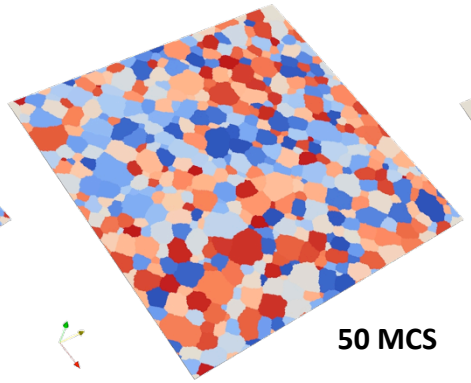
# Grain Growth (kMC)

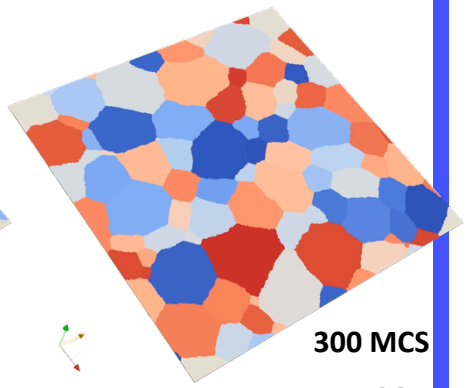



1. **Choose a site *i***
2. **Choose a new spin**
3. $$E_{GG} = \frac{1}{2}\sum_{i=1}^{N}\sum_{j=1}^{neigh}[1-\delta(q_i,q_j)]$$
4. $$P = \begin{cases} \exp\left(\frac{-\Delta E}{k_B T_S}\right) & if\ \Delta E > 0 \\ 1 & if\ \Delta E \le 0 \end{cases}$$


0 MCS

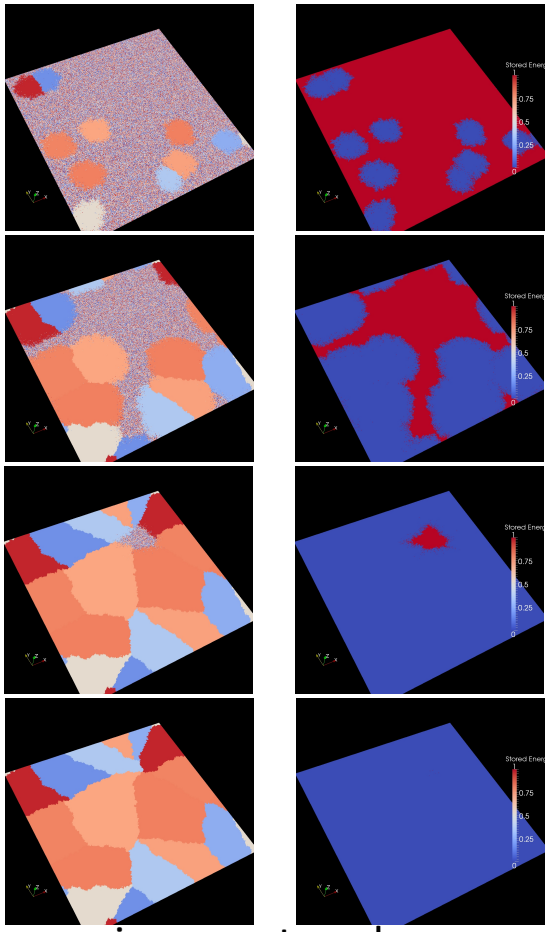

10 MCS


50 MCS


300 MCS

# Recrystallization, CA

Binary Energy Case [ Ω = 1 or 0 ]
2Dimensional, Site Saturated



spins stored energy

1. **Choose a site *i***
2. **Choose a neigh *j***

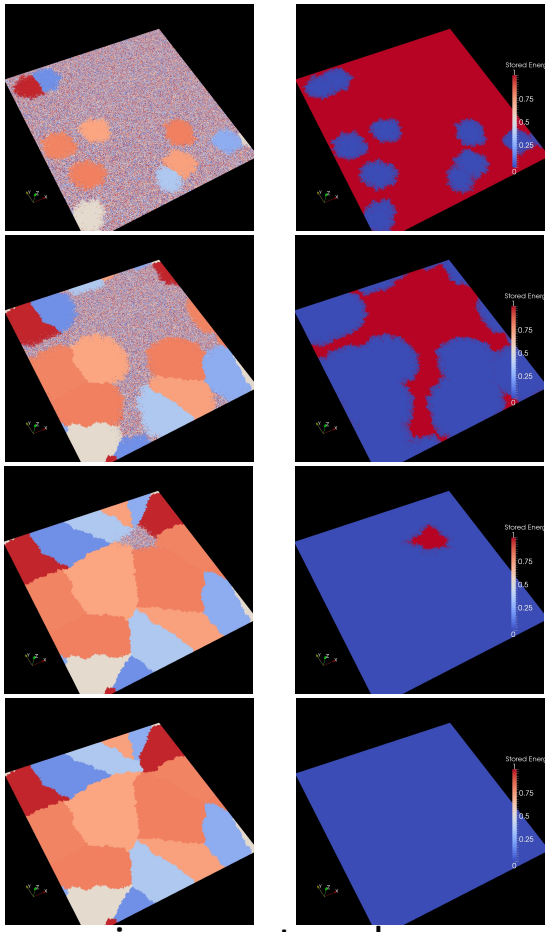
3. $$if\ \Omega_i < \Omega_j^{initial} \begin{cases} \Omega_j = \Omega_i \\ spin_j = spin_i \end{cases}$$

4. $$\Delta E_{i,RC} = \Omega_i^{final} - \Omega_i^{initial}$$

5. $$P = \begin{cases} 0 & if\ \Delta E > 0 \\ \dfrac{|\Delta E_{i,RC}|}{\Omega^{MAX}} & if\ \Delta E \le 0 \end{cases}$$

# Recrystallization, CA

Binary Energy Case [ e = 1 or 0 ]
2Dimensional, Site Saturated

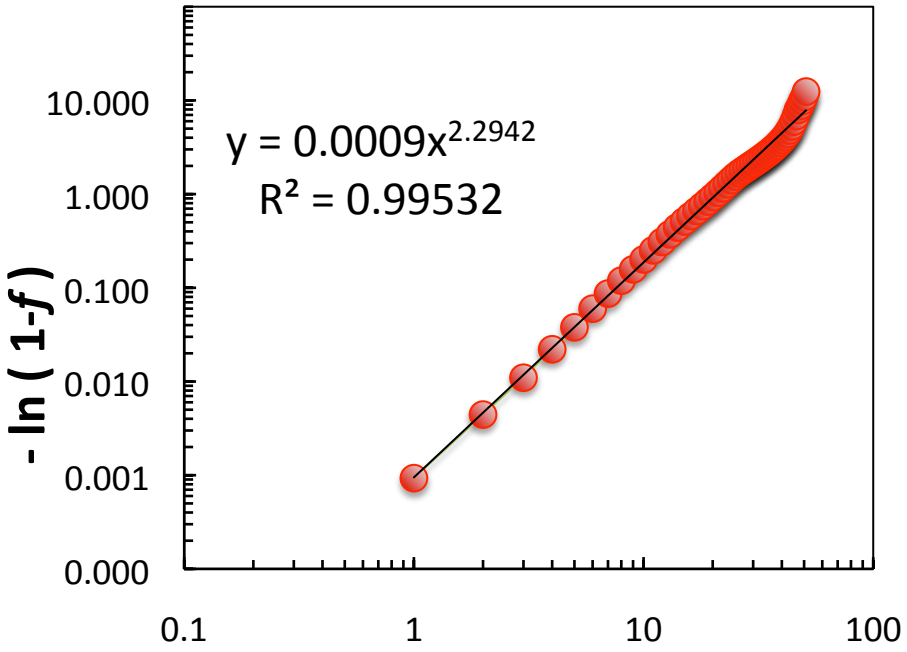


spins stored energy

$$Spins = 100$$

$$Total\ Sites = 250\,000$$

$$(1 - f) = \frac{(Total\ N_{accepted} - N_{accepted})}{Total\ N_{accepted}}$$



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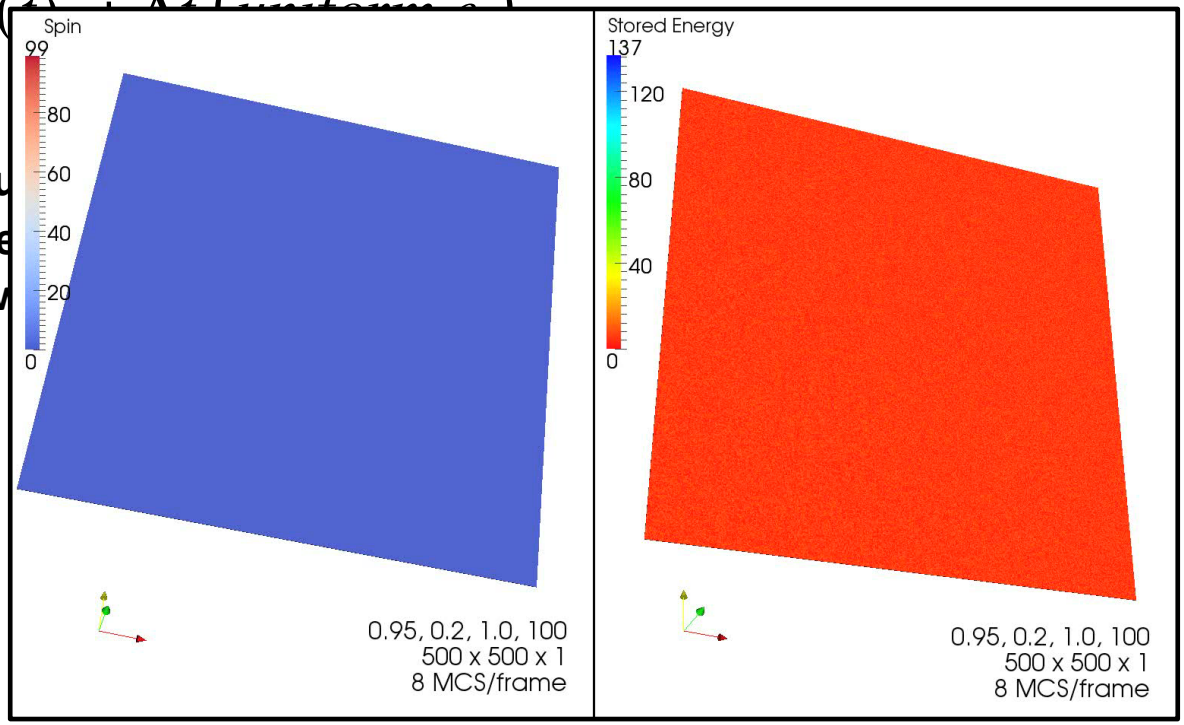
# Dynamic Recrystallization
# Energy Accumulation & Nucleation

**At each site:**

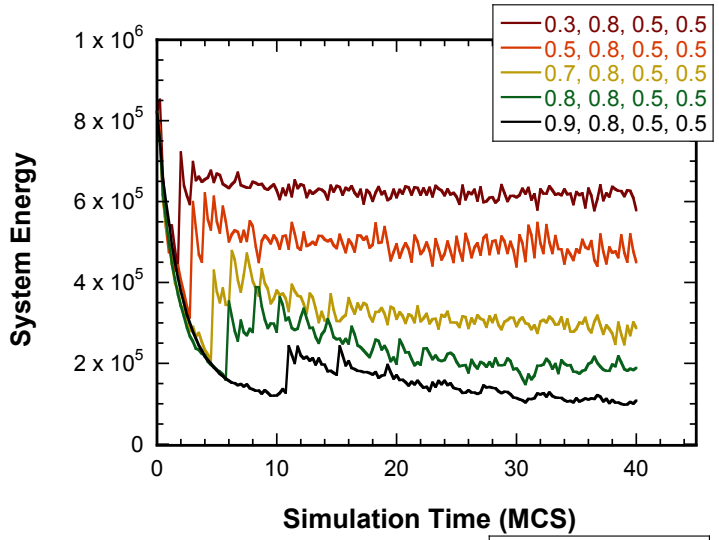
1. **Attempt GG**
2. **Attempt recrystallization**
3. **Increase stored energy by**

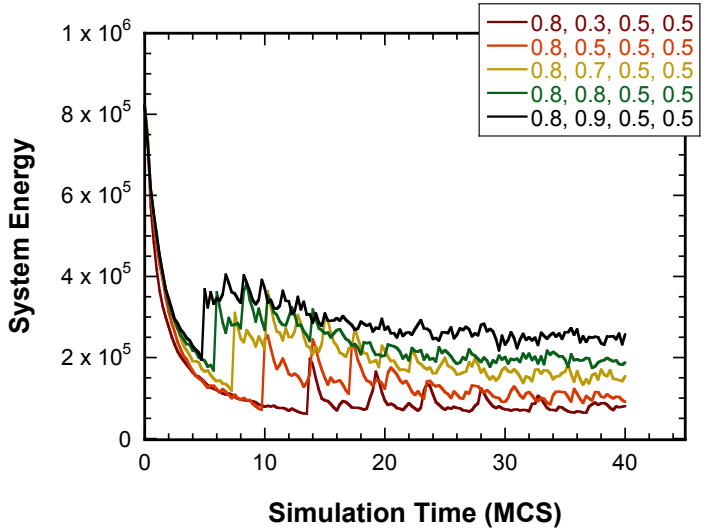
$$\Omega(i,t+1) = \Omega(t) + \Delta t\,(\text{uniform } a)$$

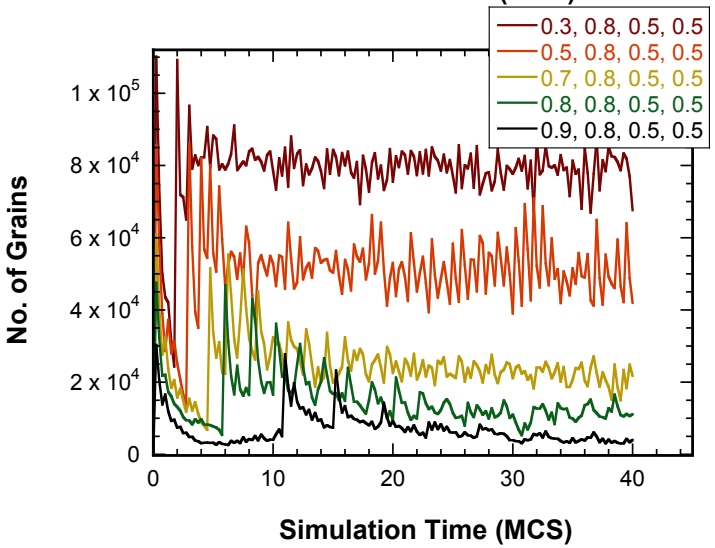
4. **Attempt nucleation u**
   - **If site stored ene**
   - **Then, nucleate v**

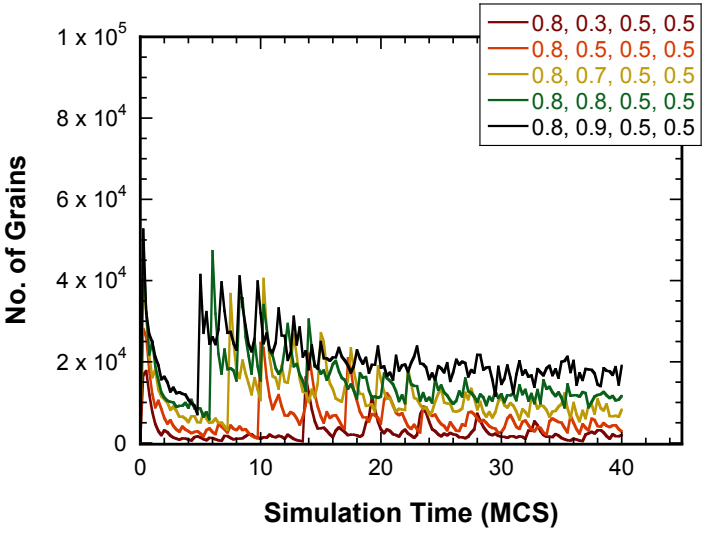


# Effects on System Energy & Grain Size




**Legend notation:** (1) threshold, (2) nucleation rate, (3) % grain growth , (4) % recrystallization steps



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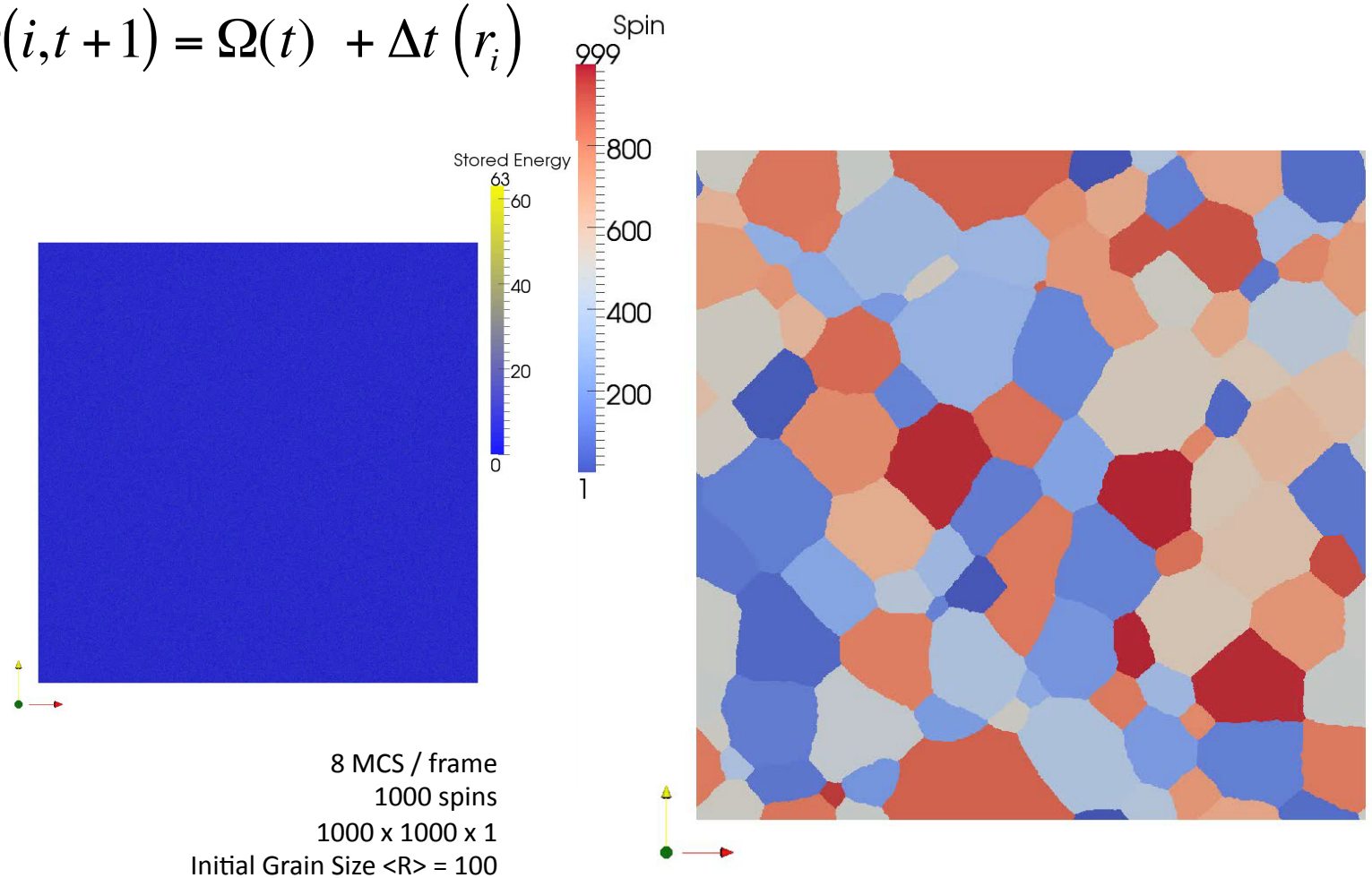
# Dynamic Recrystallization
# Hybrid Algorithm

$$\Omega(i,t+1) = \Omega(t) + \Delta t\,(r_i)$$

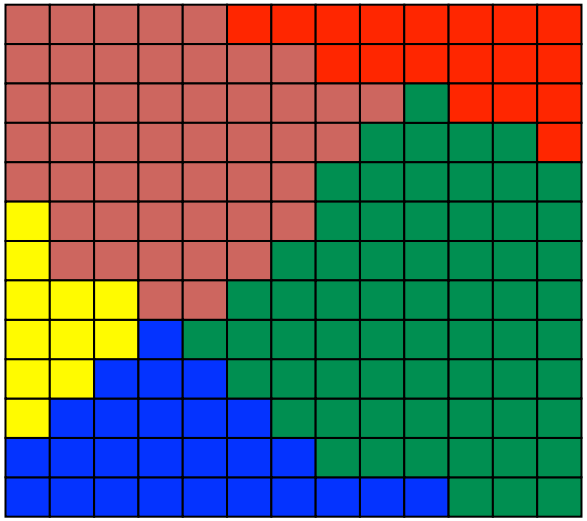



8 MCS / frame
1000 spins
1000 x 1000 x 1
Initial Grain Size <R> = 100
Average <R> During Recrystallization = 5

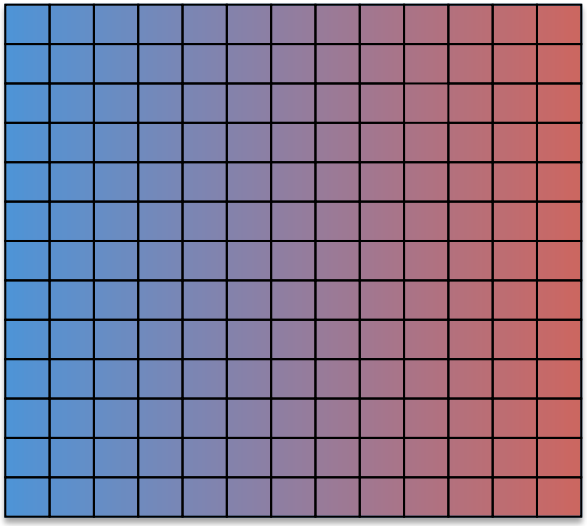
# Representation of Microstructure and Composition Hybrid Model

- 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

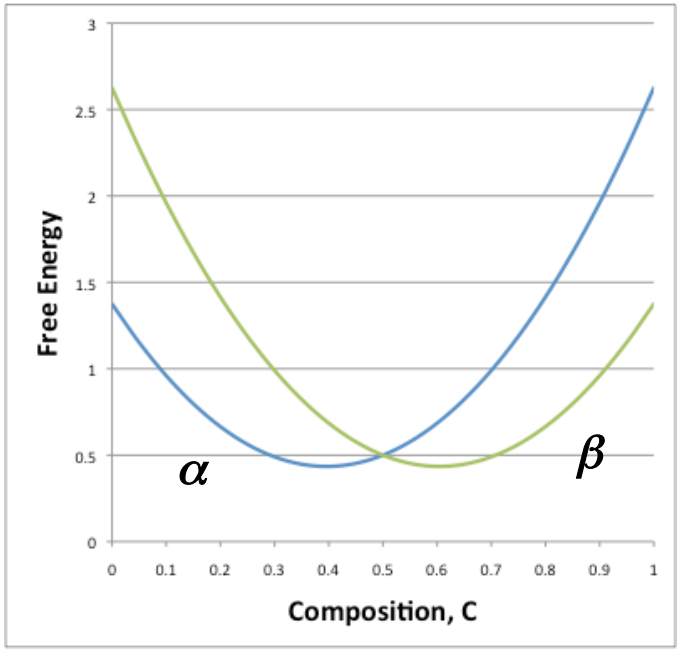
- **EOS is a function of volume free energy and interfacial energies.**

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

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

**An example of $E_v$**

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



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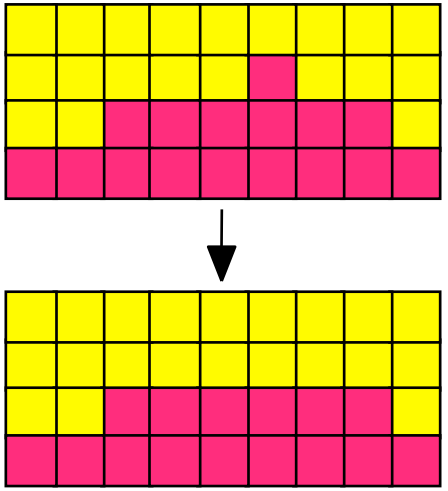
# Kinetic of Evolution Hybrid Model

- **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.**

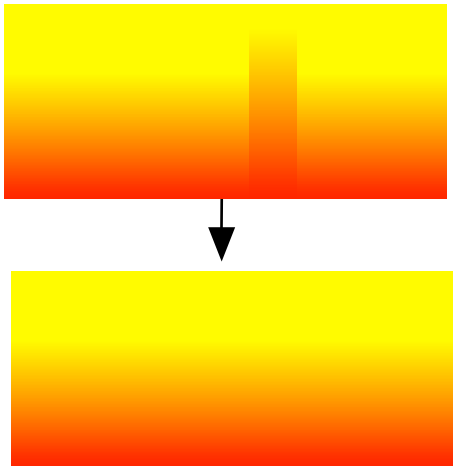
$$\frac{\partial C}{\partial t} = -M_c\left(\nabla^2 \frac{\partial E_v}{\partial C} - \kappa_c \nabla^4 C\right)$$

- **Where $E_v$ is from the hybrid Free Energy**

**grain growth change pixel color**



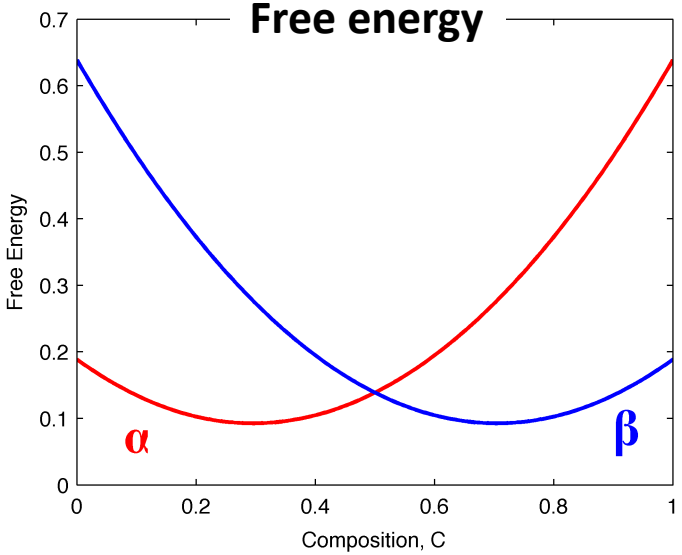
**Composition change by diffusion**

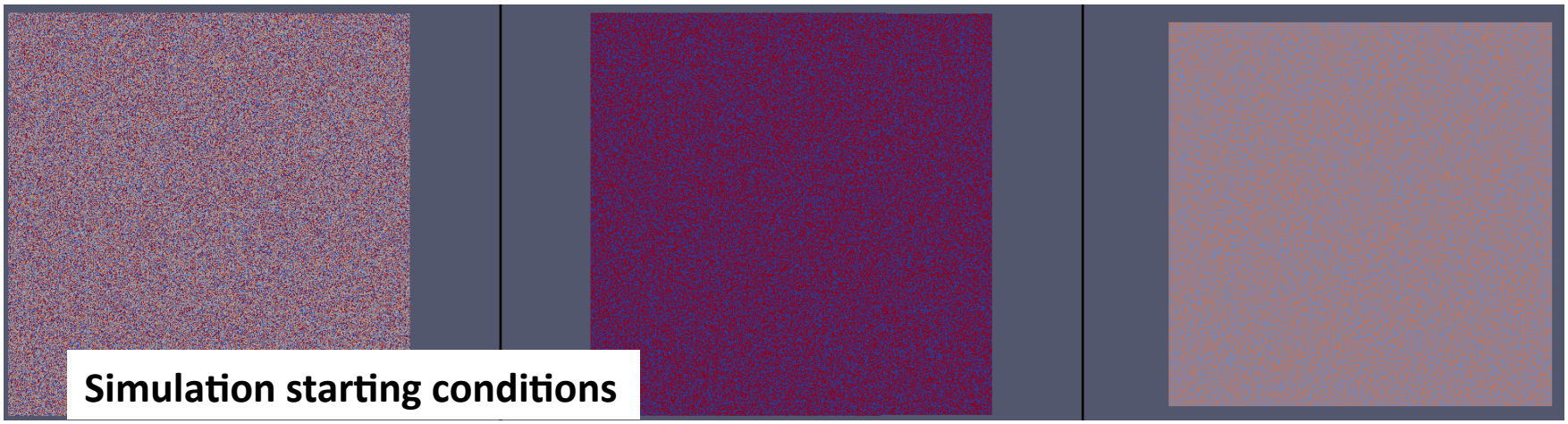


# Simulation of Grain Growth and Diffusion in a Two-Component, Two-Phase System: HYBRID Potts-PF

Coupled grain growth and diffusion simulation

- Simulation size = 1000 x 1000
- Overall composition = 0.5A and 0.5B
- Two phases = $\alpha$ and $\beta$
- Free energy curves shown
- Starting configuration – grains that are single site, $\alpha$- & $\beta$–grains of $C_\alpha$=0.25, $C_\beta$=0.75
- Time = 500 MCS (1 GG and 100 diffusion steps)



Simulation starting conditions



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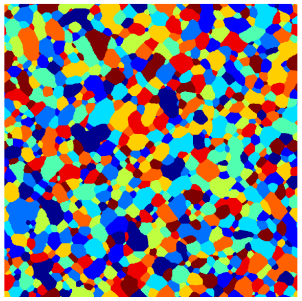
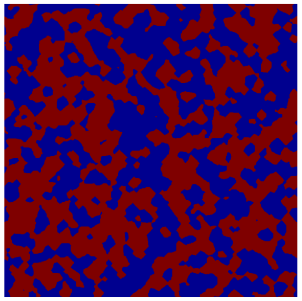
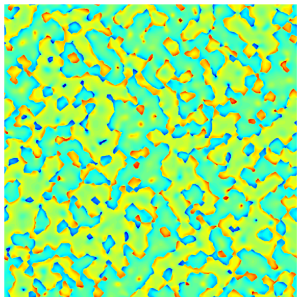
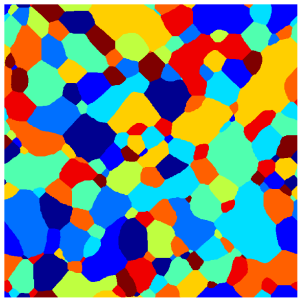
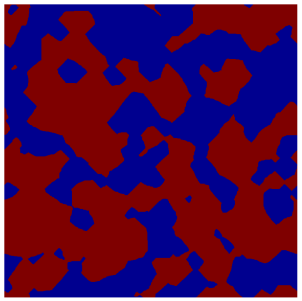
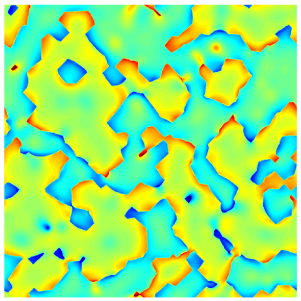
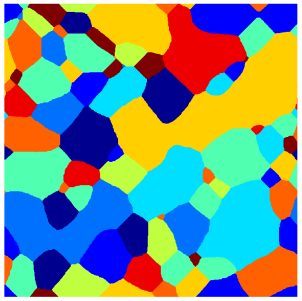
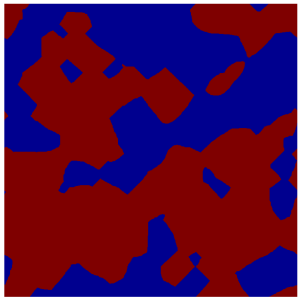
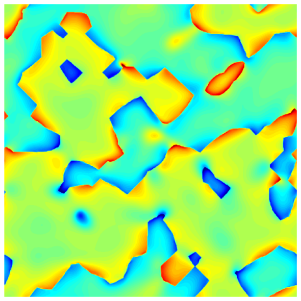
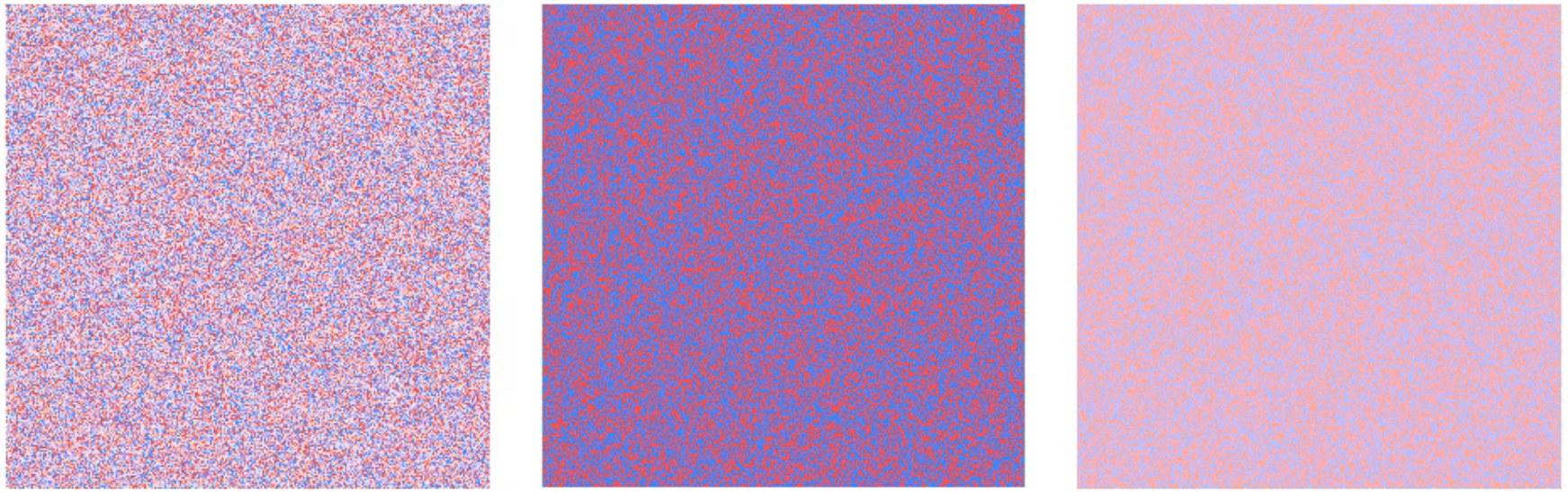
# Results of Hybrid Potts-Phase Field Model

| | Grain growth | Phase coarsening | Composition |
|---|---|---|---|
| time = 50 |  |  |  |
| 250 |  |  |  |
| 500 |  |  |  |

# Simulation of Grain Growth and Diffusion in a Two-Component, Two-Phase System: HYBRID Potts-PF

0 MCS

Grain ID | Phase | Concentration



**Simulations show:**

- **Grains grow,**
- **phase region coarsen,**
- **Composition are the minimum free energy composition in the bulk of the phase regions**

# Direction of Future Work

**We understand & describe fuel behavior at the microstructural and atomistic scales**

- **Fission gas bubbles, swelling, pore distribution**
- **O/M ratio, defects, thermal conductivity, diffusivity**

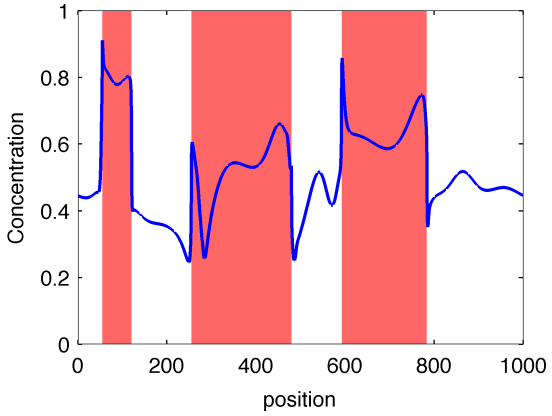
**But, we model it on the continuum scale with all these features are homogenized**

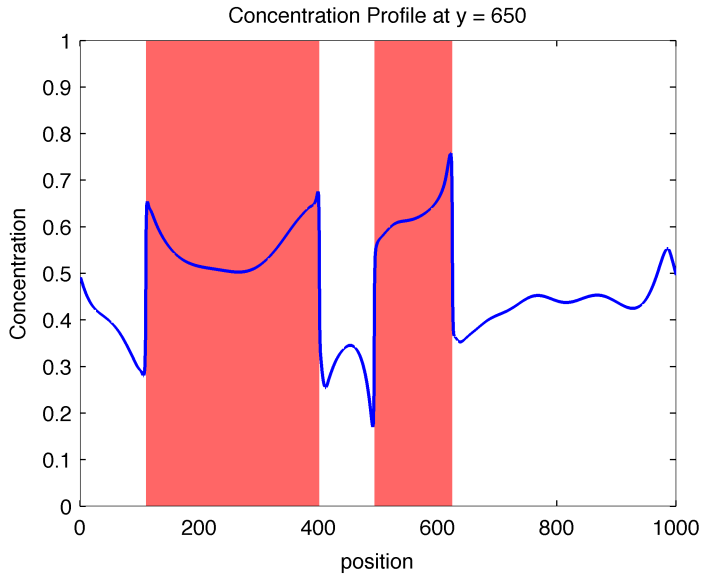
**The next step in fuel modeling is to go down to the next level of refinement – mesoscale and capture the microstructure**

- **Mesoscale models are sufficiently developed to simulate large scale problems**
- **Can couple to atomistic and continuum**
- **Enable understanding (and prediction) on this scale**
- **Enable designing and engineering fuel at the scale.**

**Hybrid mesoscale models that can integrate all the necessary physics for true microstructural evolution modeling is the future.**

# Simulation of Grain Growth and Diffusion in a Two-Component, Two-Phase System: HYBRID Potts-PF






Sandia National Laboratories