Meso–Scale Modeling of Fission Gas Bubble Induced Swelling

SAND2013-9185C

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Outline

Introduction
- Experimental Motivation
- Previous Work

Model Development
- Simulation Framework
- Modeling FG Bubble Swelling
- System Thermodynamics

Results and Analysis
- Microstructural Evolution
- FG Release by Percolation
- Comparison Between Initial Porosities
- Nano–fibrous Structures

Conclusion

Release of Gaseous Fission Products

- Xe and Kr are the main gaseous fission products
  - Relatively high yields
  - Low solubility in fuel
- Nucleates and precipitates as bubbles
  - Both, inter– and intragranular
- Fission gas bubbles leads to swelling
  - Degrade gap conductivity
  - Clad ballooning and burst
  - Number and size distribution of bubbles
- Build up can lower reactivity, i.e. xenon poisoning
  - Neutron absorber
- Understanding release mechanisms
  - Design better (safety and efficiency) fuels
  - Achieve higher burnups

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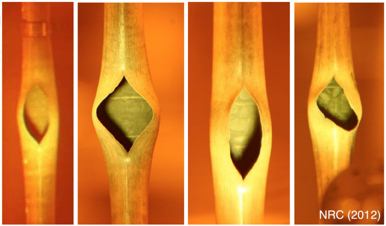

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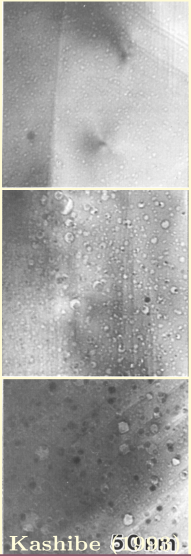
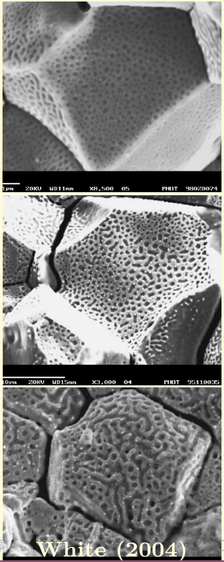
Accumulation and Release of FG

Intragranular bubbles



Intergranular bubbles



Fission gas within fuel, can be:

- Dissolved in fuel matrix (lattice)
- Form intragranular nano–bubbles
- Form intergranular micro–bubbles

And can be released by:

- Direct release from fission and knocked–out
- Diffusion to the surface
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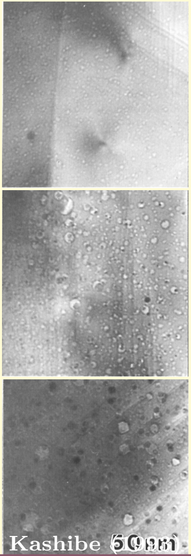

Kashibe (1993)

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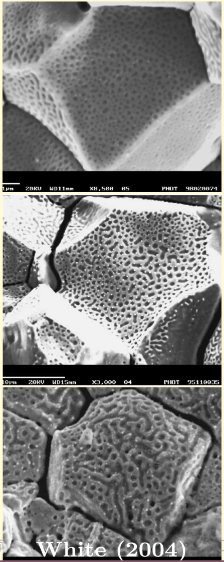
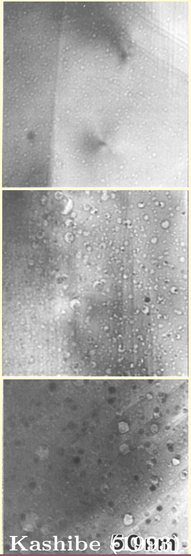


White (2004)

Accumulation and Release of FG

Intragranular bubbles



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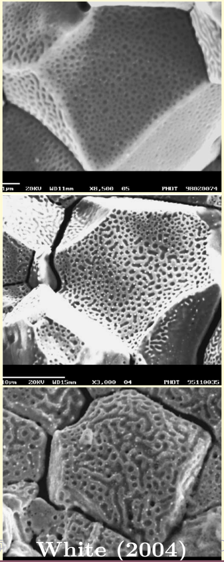

Swelling Due to Gaseous FP Bubbles

- Main difference between a void, concentration of FG
  - In TEM, differentiated by the feature shape (faceted or sphere)
- At high temperatures, fission gas diffuses into bubbles
  - Grows as voids reach bubble
  - Reaches equilibrium with surface energy as FG reaches bubble
- For a distribution of bubbles

$$\frac{\Delta V}{V} = \frac{\left(\frac{4\pi}{3}\right)\sum R_i^3 N_i}{1 - \left(\frac{4\pi}{3}\right)\sum R_i^3 N_i}$$

- Can lead to fuel–clad interactions

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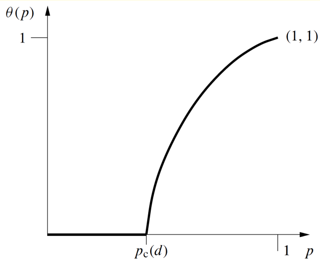
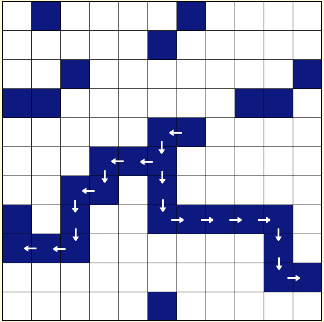
Fission Gas Release: Percolation Theory

- Behavior of connected clusters in a random graph
  - Pour liquid from top
- Simulate FG release through grain boundary



Grimmett (1999)



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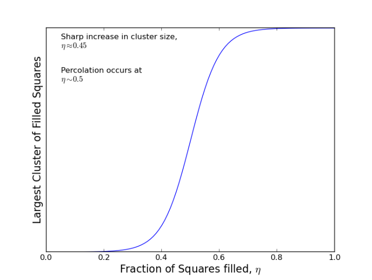
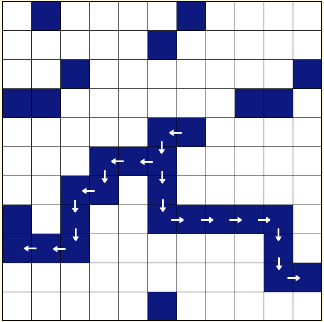
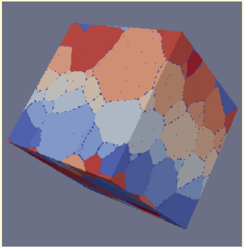




Grain Boundary Bubble Percolation

- Previous work (*Tikare*), applied percolation model to FG release
- Potts kMC model simulated microstructural evolution
  - With similar FG release percolation model
- Primary gas release mechanism is intergranular bubbles percolation
  - Percolating feature releases gas to free surface (external or crack)
  - Intergranular bubble formation is complex

Gas bubbles in polycrystal



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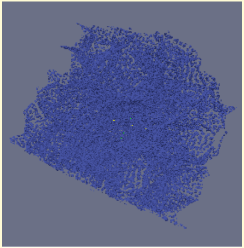
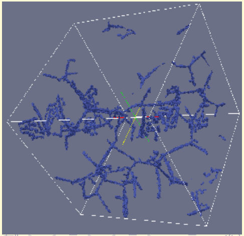


Percolating bubble



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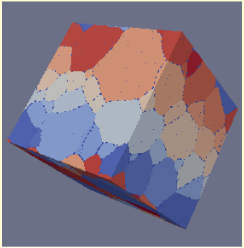

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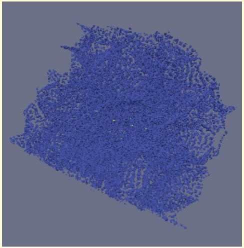

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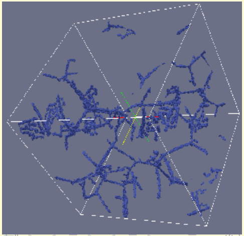

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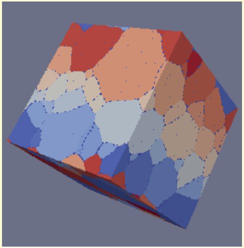

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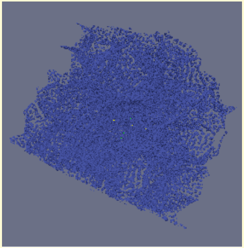

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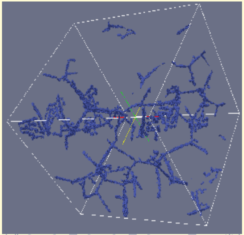

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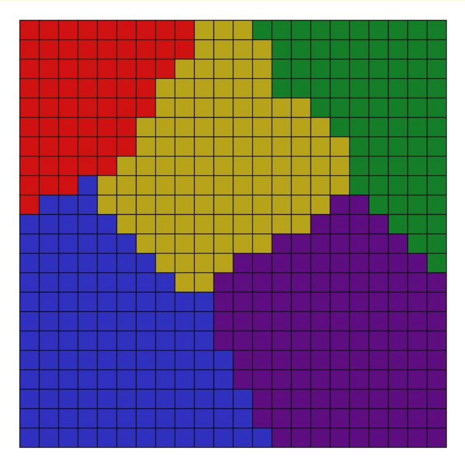
Microstructural Representation



- 3D digitized microstricture
- Each color is a different phase/state
  - Solid grain (orientation)
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  - Vacuum/void
- Interfaces, grain boundaries & bubble surfaces are surface between different "colored particles"

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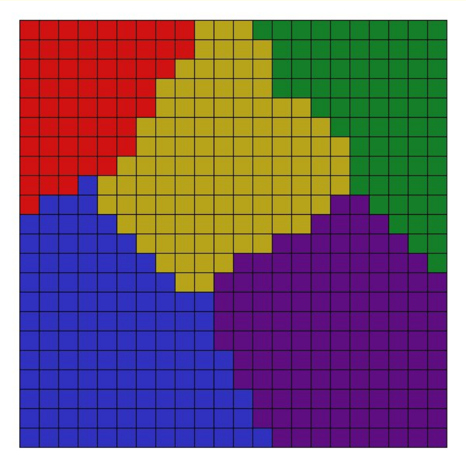

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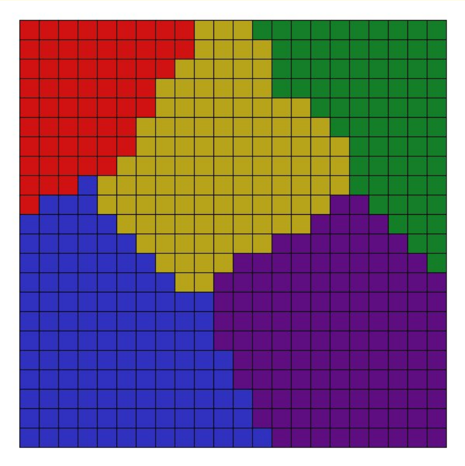

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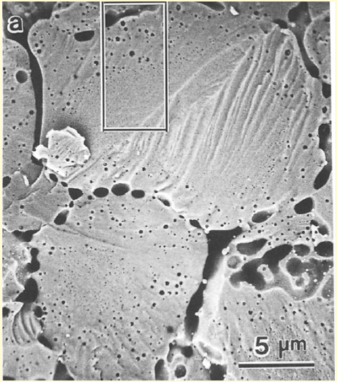
Bubble Nucleation



White (2004)

- Gases are formed from fission and transmutation events
- Different "paths" to follow
  - Direct release
  - Nucleation and growth of bubble
  - Resolution into the matrix
  - Etc.
- We considered bubbles that have migrated to GB
  - These nucleateat a given frequency
  - Once at the GB, they coalense into large bubble structures

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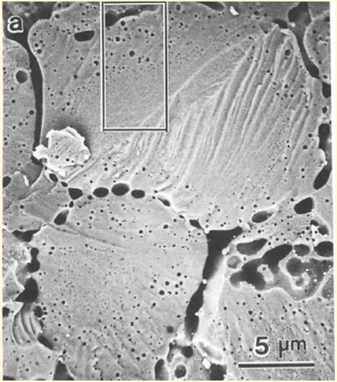

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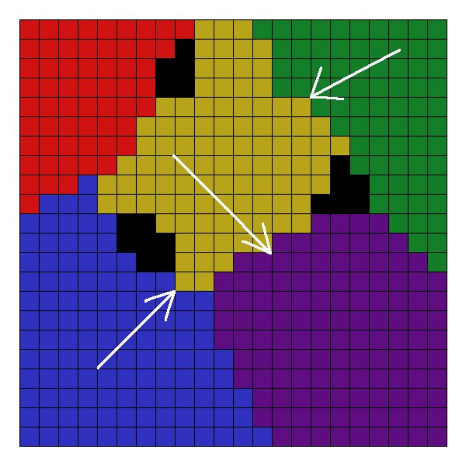

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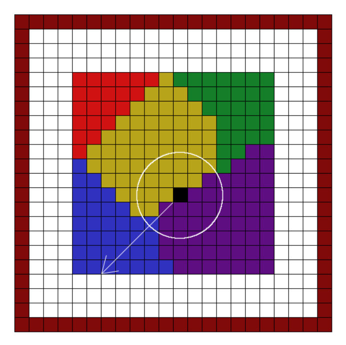
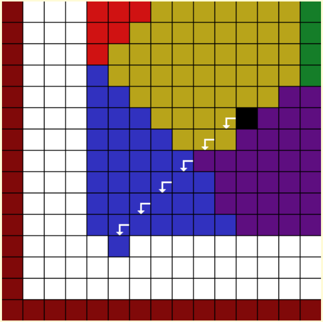
Bulk Volumetric Swelling





- Swelling simulated by a site exchange mechanism
- Choosing a random direction

$$p_x = \cos(\phi)\sin(\theta) \qquad p_y = \sin(\phi)\sin(\theta) \qquad p_z = \cos(\theta)$$

- Essentially, "reverse" sintering [*García–Cardona, Tikare* (2011)]

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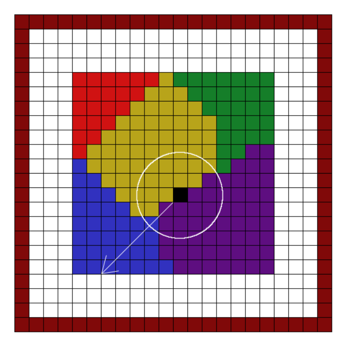

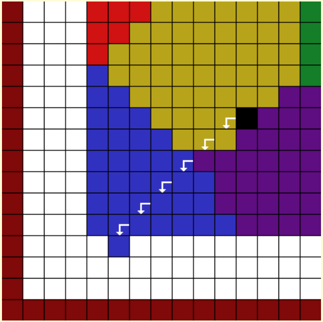

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Equations of State and Kinetics

- EOS defines energy of system

$$E = \underbrace{\sum_{i=1}^{N} \left(D_{i=ts} + G_{i=dg}\right)}_{\substack{\text{Volumetric} \\ \text{energy terms}}} + \underbrace{\sum_{i=1}^{N} \sum_{j=1}^{n} \alpha_i \left(1 - \delta_i\right)}_{\substack{\text{Interfacial} \\ \text{energy terms}}}$$

- Evolution simulated by statistical rearrangement of ensemble
  - Energy change used to determine thermodynamics of the change
- To determine the probability of a kinetic event taking place

$$p_{event} = \frac{r_{event}}{n_{sites}\omega} = \underbrace{\exp\left(-\frac{\Delta E}{k_B T}\right)}_{\text{Boltzmann Distribution}}$$

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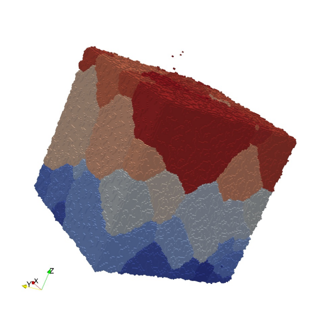
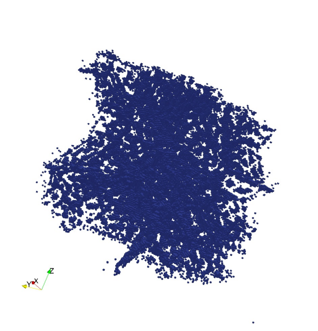
Microstructural and Volumetric Changes

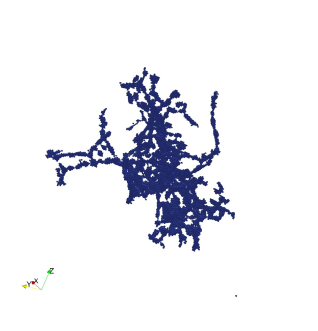
Swollen microstructure



Gas bubbles



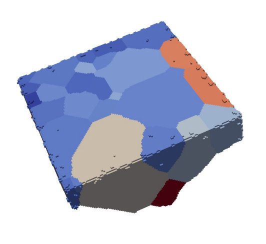
Percolating bubble



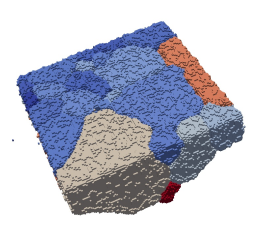
- Percolation starts when bubble density is $\sim 3\%$
- Percolating bubble is $\sim 30\%$ of bubble volume

Stages of Percolation

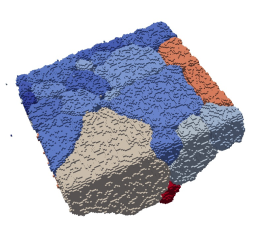
Initial Microstructure (time = 0 MCS)



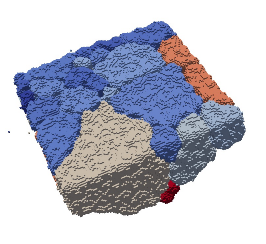
Early Stage (5503 MCS)



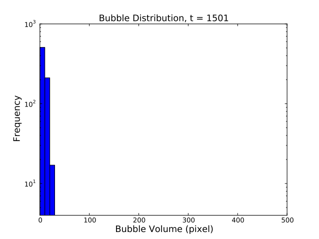
Moderate Stage (9000 MCS)

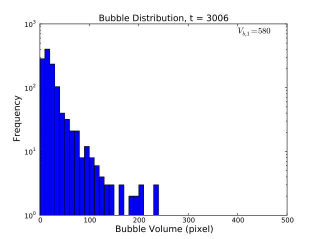


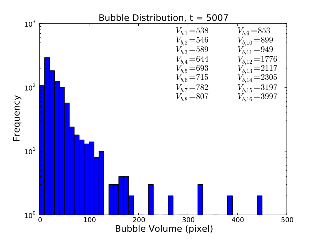
Advanced Stage (24006 MCS)

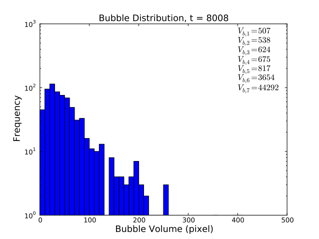


Gas Bubble Distribution

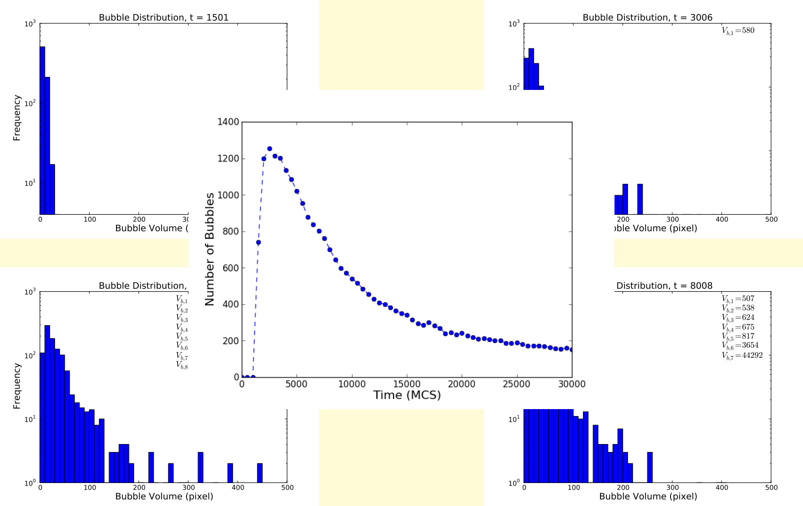




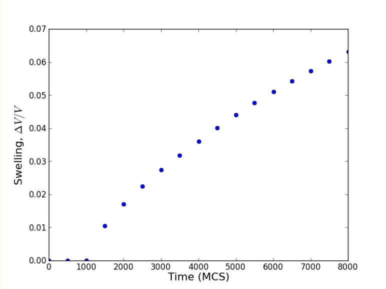


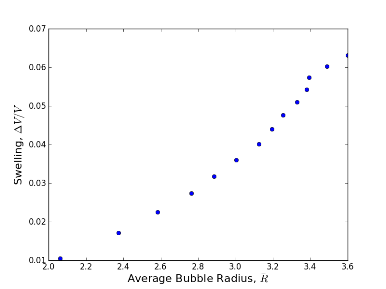


Gas Bubble Distribution

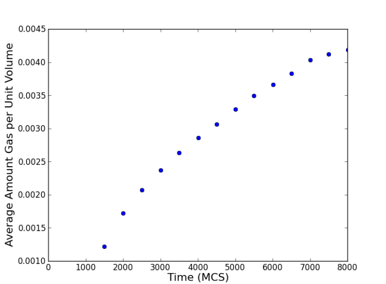


Gas Bubble Swelling

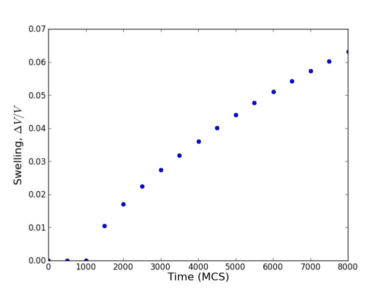


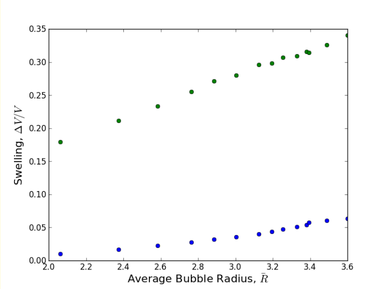


$$\frac{\Delta V}{V} \approx \frac{4\pi R_b^3}{3} N_b$$



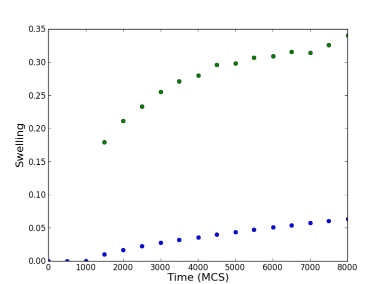
Gas Bubble Swelling



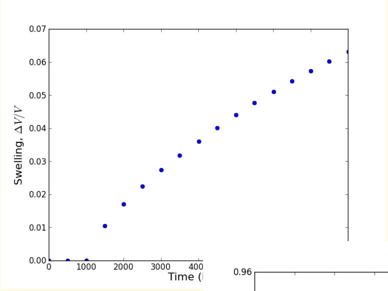


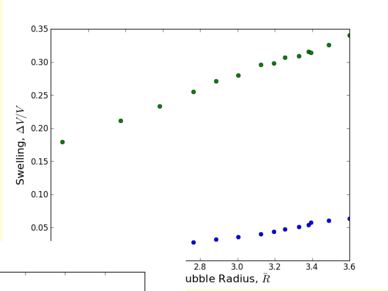
$$\frac{\Delta V}{V} = \frac{2(R_{gb}/a)(R_{gb}/\mathcal{R})^2}{1 - 2(R_{gb}/a)(R_{gb}/\mathcal{R})^2}$$

Agrees with *Kagana*,
$\sim 83\%$ lower swelling for interlinkage



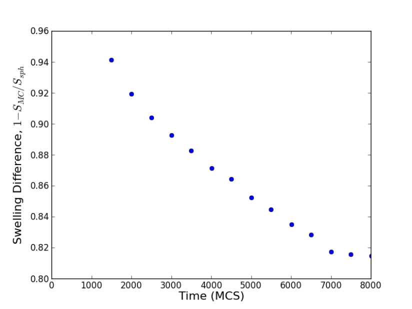
Gas Bubble Swelling

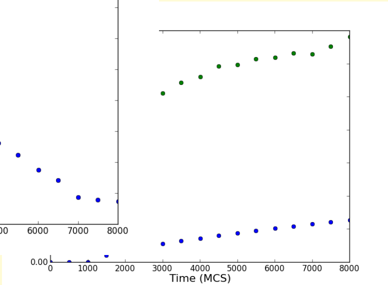




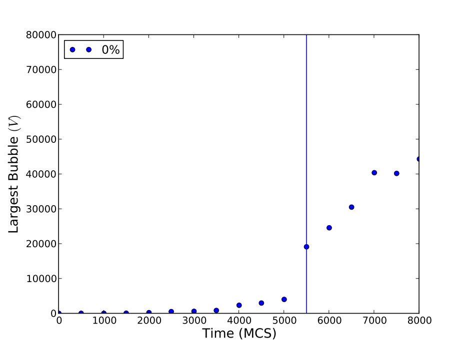
$$\frac{\Delta V}{V} = \frac{2(R_{gb}/a)}{1 - 2(R_{gb}/}$$

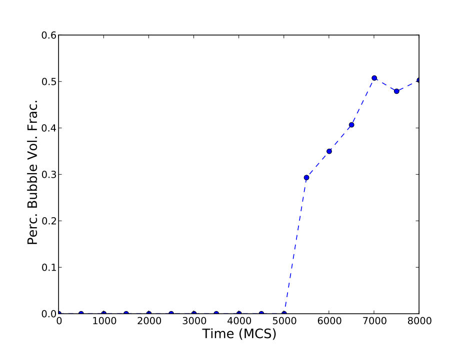
Agrees with *Kagana*,
$\sim 83\%$ lower swelling
interlinkage





Bubble Evolution





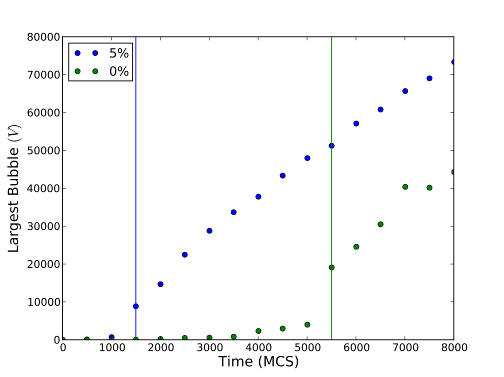
- Substantial increase in bubble volume
- Abrupt increase in volume at $t = t_{perc} \approx 5500$

Initial Porosity of 5%

- Initial porosity has significant consequences
  - Percolation time significantly reduced

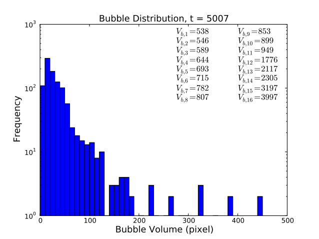
$$\frac{t_{perc}^{\rho=95\%}}{t_{perc}^{\rho=100\%}} \sim 27\%$$

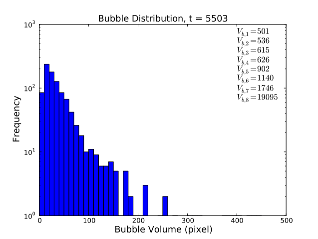
  - Percolation happens with smaller bubbles

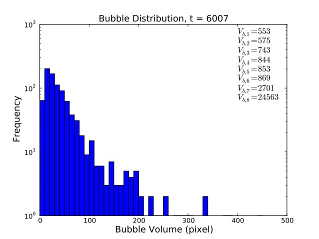


Porous: Gas Bubble Distribution

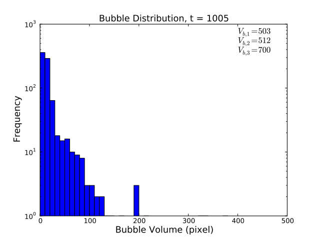
Porosity = 0%

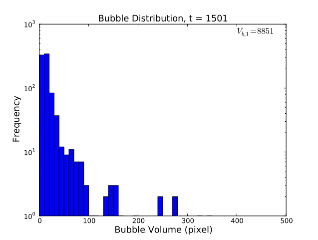


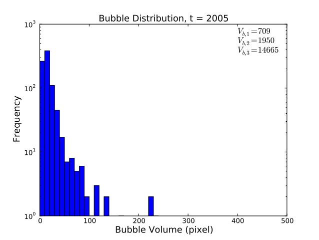




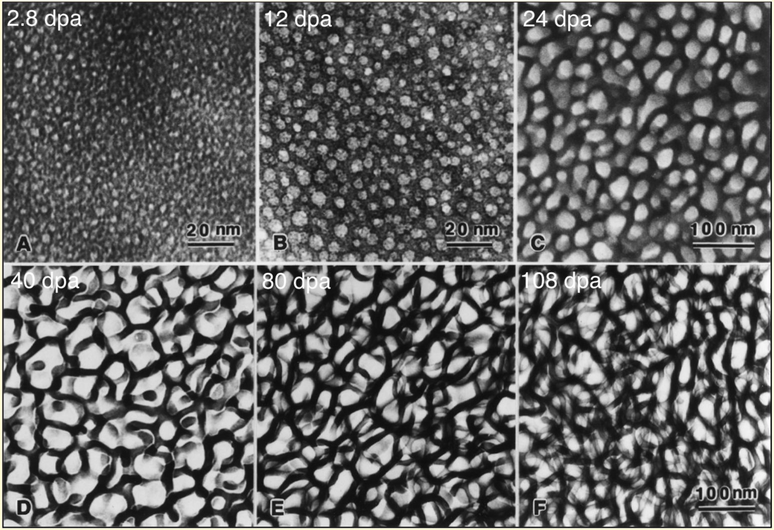
Porosity = 5%



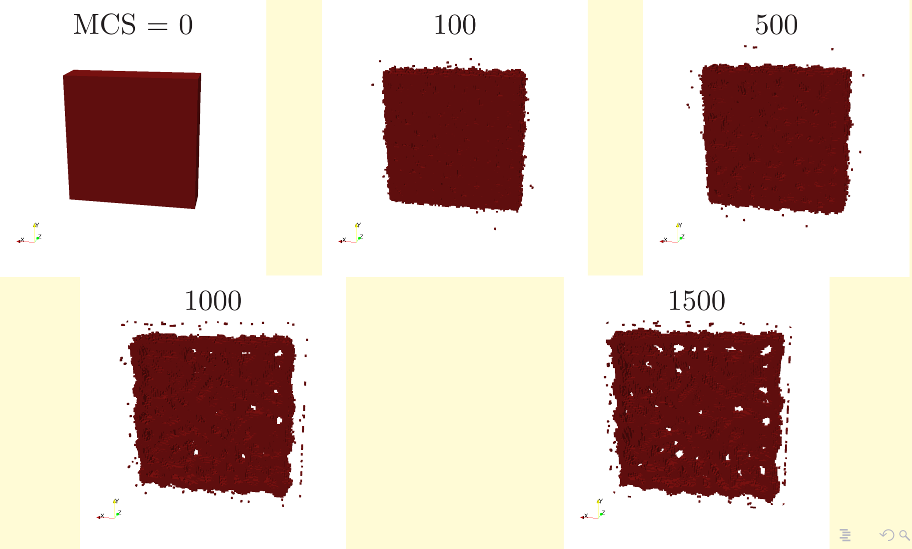




Self-assembled Nano-porous Structures I



Self-assembled Nano-porous Structures II



Outline

Introduction
- Experimental Motivation
- Previous Work

Model Development
- Simulation Framework
- Modeling FG Bubble Swelling
- System Thermodynamics

Results and Analysis
- Microstructural Evolution
- FG Release by Percolation
- Comparison Between Initial Porosities
- Nano–fibrous Structures

Conclusion

Summary

- Monte Carlo swelling model is able to simulate microstructural evolution of bubble gas swollen fuels
- Intergranular bubble formation and interlinkage is a complex behavior
- Percolation depends on microstructure, e.g. grain size
  - It also seems like volumetric swelling has an effect on percolation
- Simple swelling model seems suitable to simulate extreme volumetric swelling, leading to high porosity nano–porous structures
- Further parameterization studies needed to fully understand the capabilities of the model

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Rackham Graduate School of UM
Sandia National Laboratory

References I

[1] Olander, D.R., *Fundamental Aspects of Nuclear Reactor Fuel Elements*, Technical Information Center, TID-26711-P1 (1976).

[2] Kashibe, S., Une, K. and Nogita, K., *Formation and growth of intragranular fission gas bubbles in UO_2 fuels with burnup of 6–83 GWd/t*, J. Nuc. Mats., v. 206, p. 22 (1993).

[3] Grimmett, G., *Percolation*, Second edition, Springer-Verlag, New York, (1999).

[4] White, R.J., *The development of grain-face porosity in irradiated oxide fuel*, J. Nuc. Mats., v. 325, p. 61 (2004).

[5] G. Was, *Fundamentals of Radiation Materials Science: Metals and Alloys*, first ed., Springer, New York (2007).

[6] Kagana, G. and rest, J. *A Physical Description of Fission Product Behavior in Fuels for Advanced Power Reactors*, ANL report, ANL–07/24 (2007).

References II

[7] García–Cardona, C., Tikare, V. and Plimpton, S.J., *Parallel Simulation of 3D Sintering*, Int. J. Comput. Mats. Sci. & Surf. Eng., v. 4, p. 37 (2011).

[8] Flanagan, M. (preparer), *Mechanical Behavior of Ballooned and Ruptured Cladding*, NRC report, NUREG-2119 (2012).

Thanks!

Questions?