Mesoscale Modeling of Nuclear Fuel Microstructures

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Nuclear Energy

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Outline

1. Overview of Fuel Conditions and Microstructure

- *Fresh fuel characteristics*
- *Microstructure evolution in reactor (swelling, gas kinetics, rim structure)*

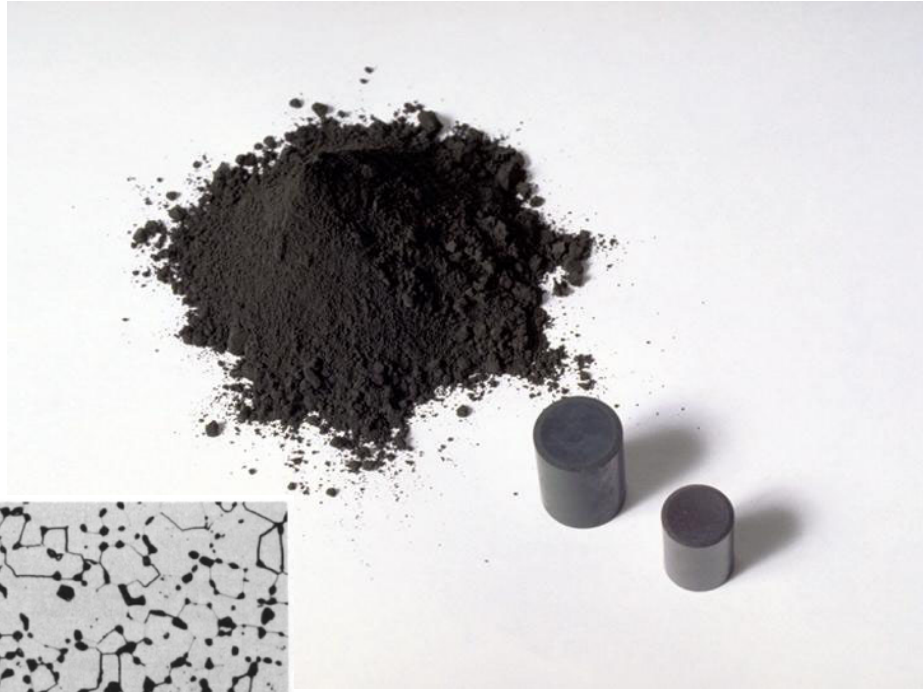
2. INL Modeling Approach to Fission Gas Evolution in Fuel

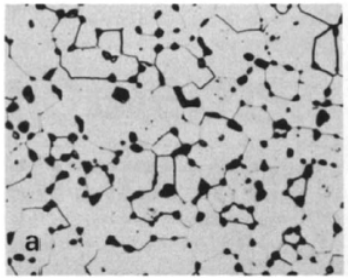
- *Bubble nucleation (Monte Carlo approach)*
- *Growth and percolation (Phase-Field method)*
- *Gas venting from fuel (Percolation method)*

3. MARMOT

- *Multi-scale microstructural modeling into continuum codes (MOOSE-BISON)*
- *Experimental measurements/scans uploaded into simulations*

Fresh Fuel

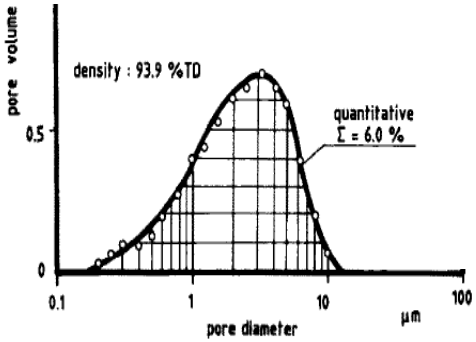


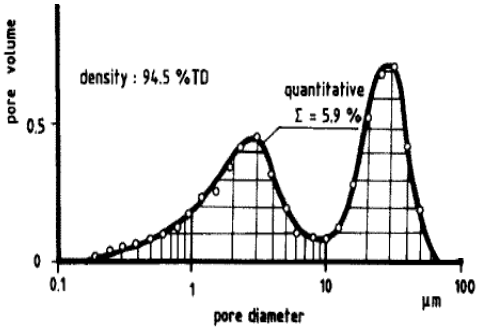


Fabrication: powder-compact, sintering

Dimensions: diameter (~ 1 cm), height (~ 2 cm)

Enrichment: ~ 5 % (U^{235})





Hodkin, JNM 88 (1980) 7.

Grain Size: ~ 10 µm

Pore Size: ~ 3 µm (uni-modal)

~ 3 µm & 20 µm (bi-modal)

Density: ~ 95%

Fuel Microstructure

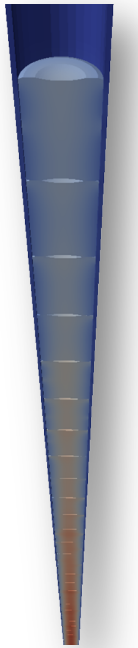
Physical Conditions

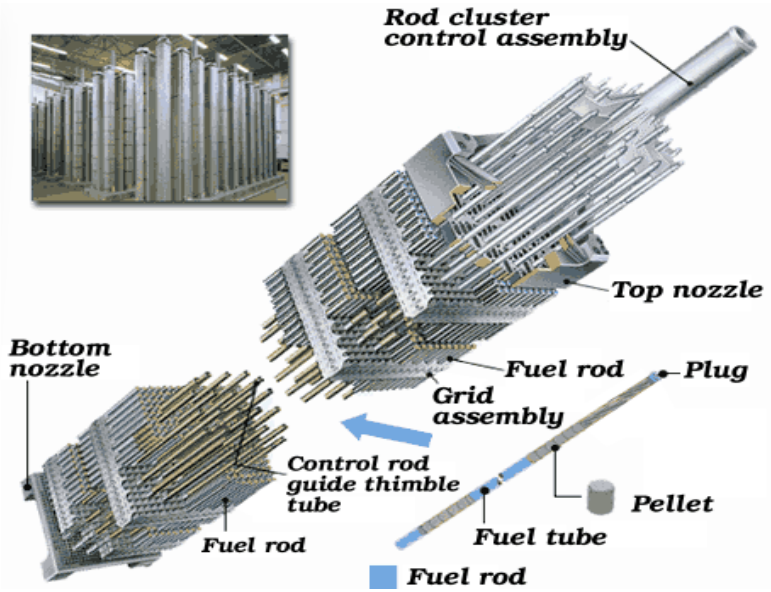
Temperatures: 400° – 1200° C
(ΔT = 1000° C / cm)

Damage Rate: 10^{-10} – 10^{-6} dpa/s

Stresses (approximate): ??

Fuel lifetime (in reactor): ~ 1 year



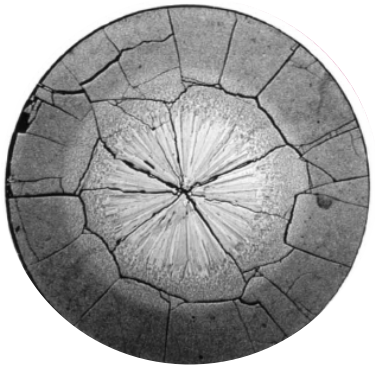


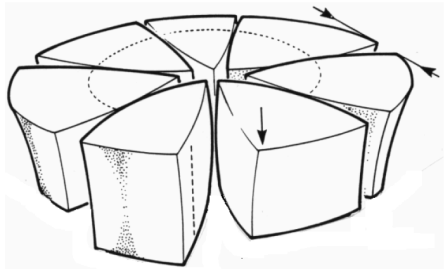
Fuel Swelling/Cracking

Non-uniform swelling due to temp. grad.

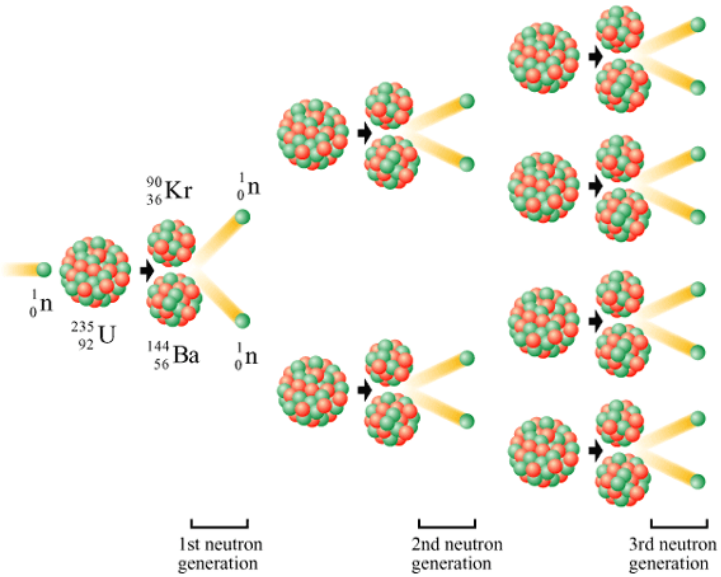
Radial cracks that form 'pie' slices

Occurs during initial power-up…





Fuel Microstructure

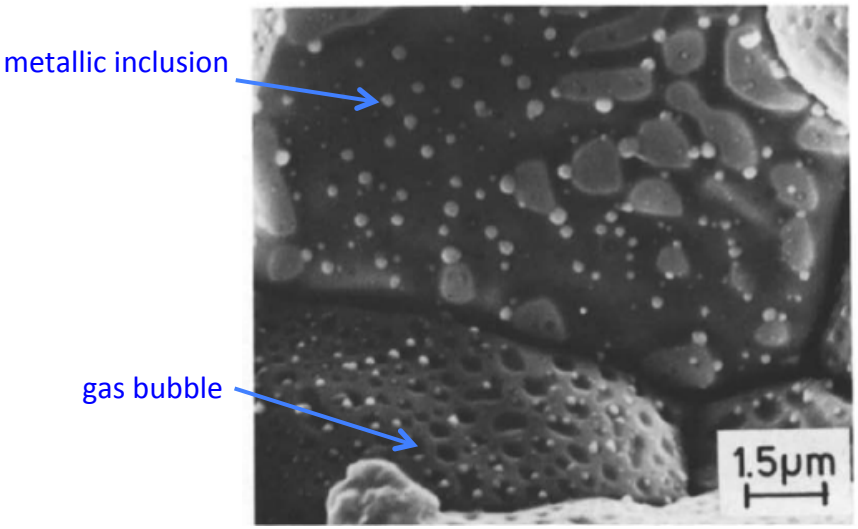


Fission Products

Solid: Ba, Zr, Mo, Cs, Sr, Ru, Nb, ….

Gaseous: Kr, Xe

Importance…? fission products: (1) produce swelling, (2) change oxygen potential, (3) release gas {pressure on cladding}.

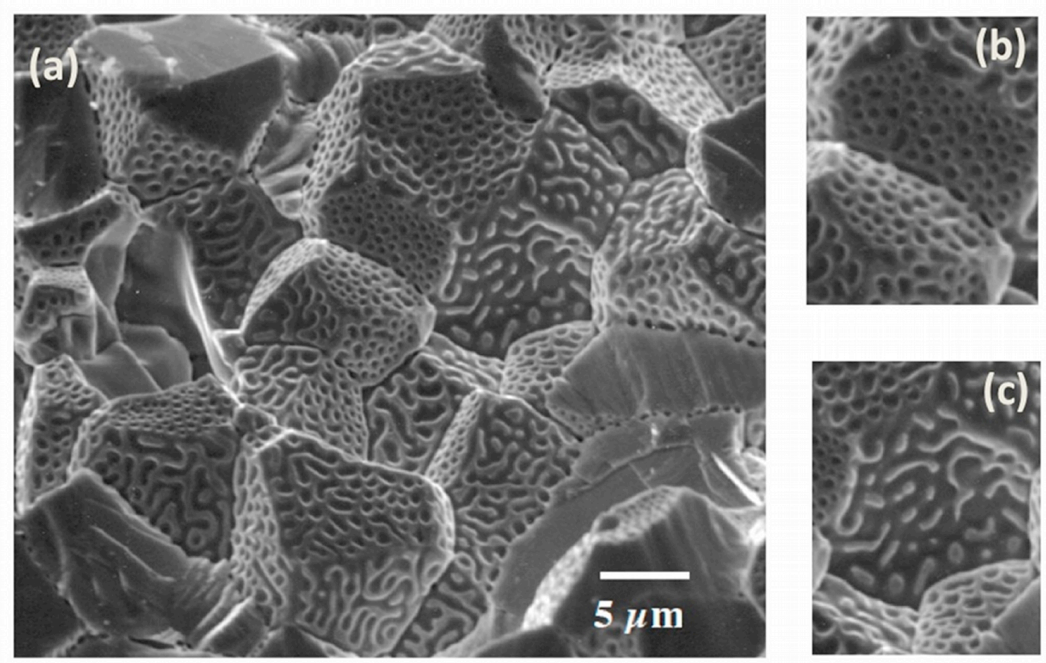


Walker et al., JNM 160 (1988) 10.

Precipitation

- Gas bubbles
- Metallic inclusions
- Oxide inclusions

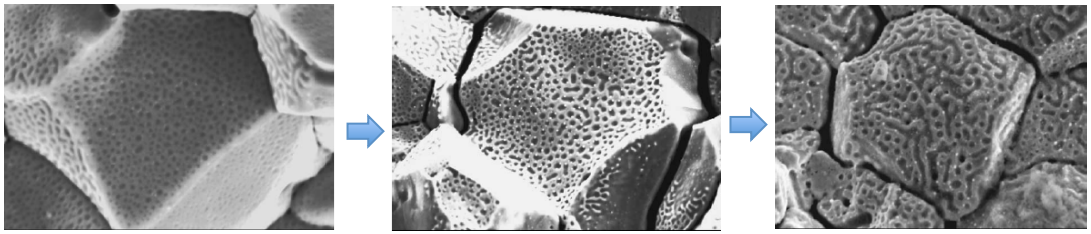
Fuel Microstructure



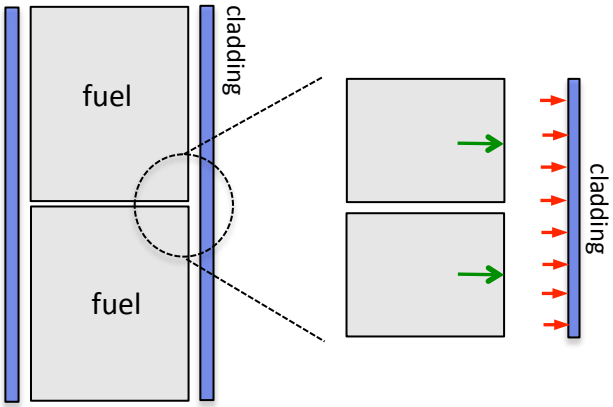
Valin et al. NEA Proc. 2000

Intergranular Gas Bubbles

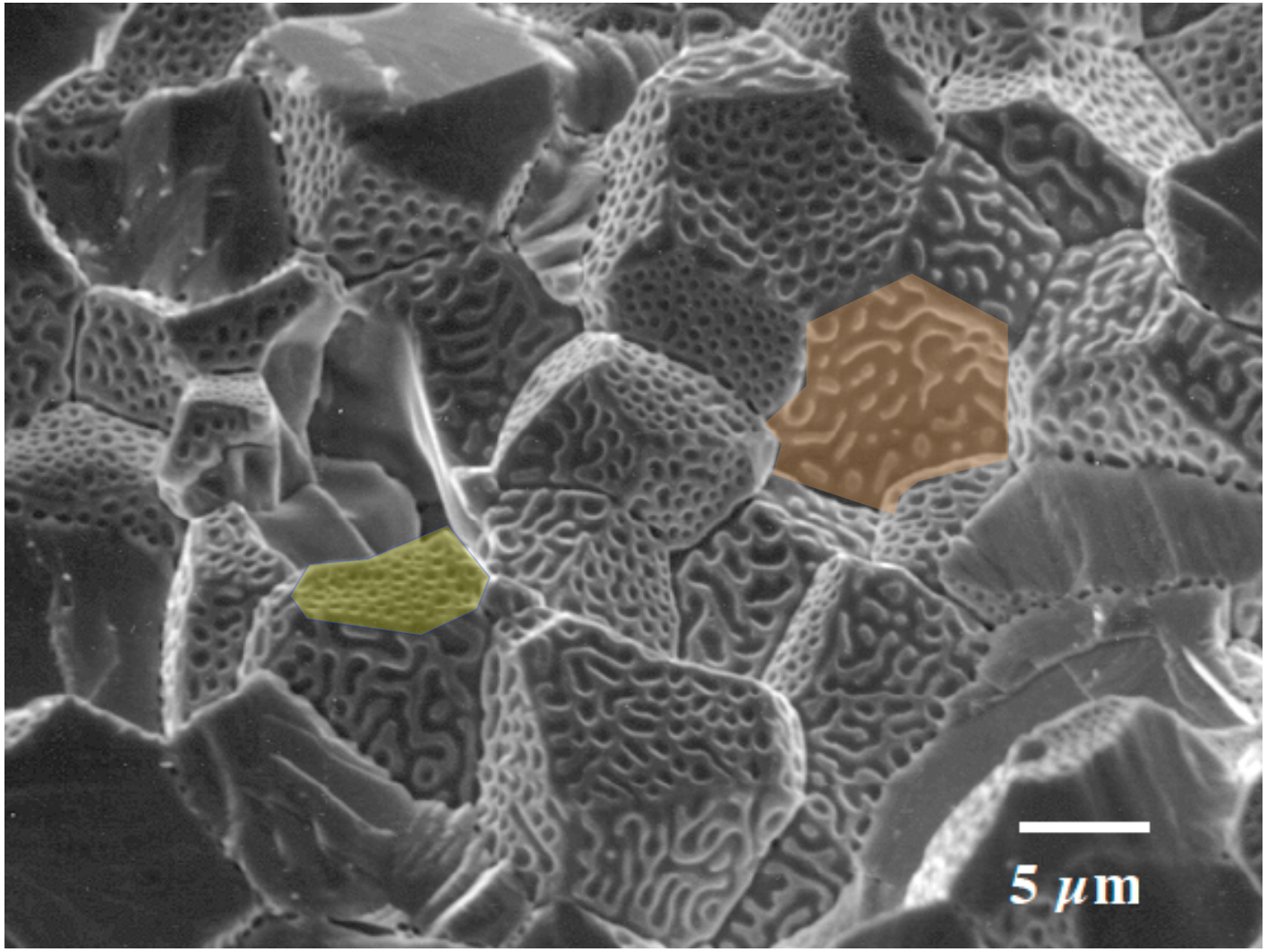
- Large bubble growth on grain boundaries
- Bubble percolation major mechanism of fission-gas release
- Bubble structure varies from GB to GB!



Nucleation Growth Percolation

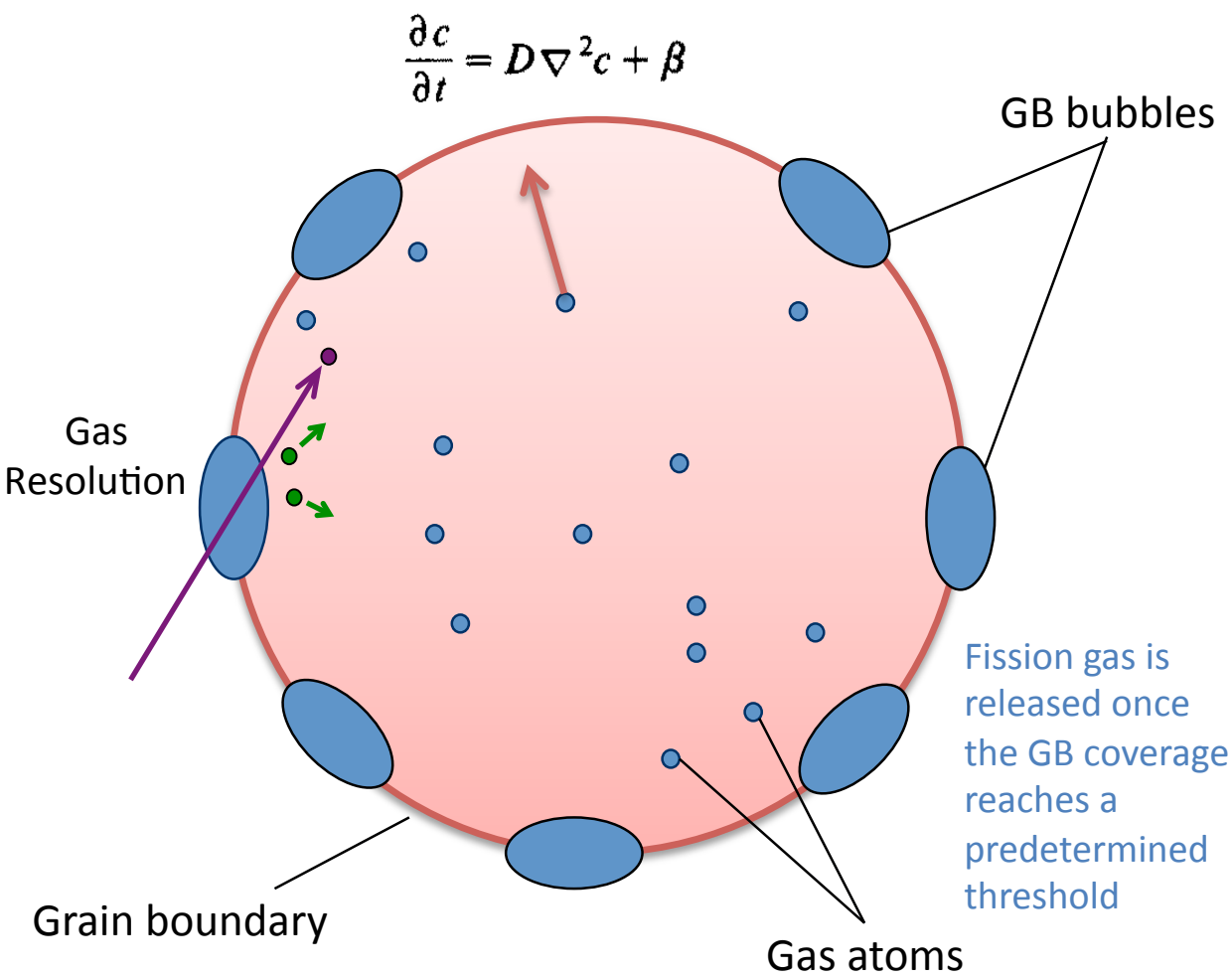


Fission Gas Release



Why do some grain boundaries have percolated bubble structures, while others do not?

Traditional Model



Fission gas is released once the GB coverage reaches a predetermined threshold

Stage 1:

- Effective gas flux to GB

- Fick's law diffusion:

$$\frac{dN}{dt} = 2\beta\sqrt{\frac{4Dt}{\pi}}\left[1 - \frac{b\delta N}{2D\beta t}\right]$$

Stage 2:

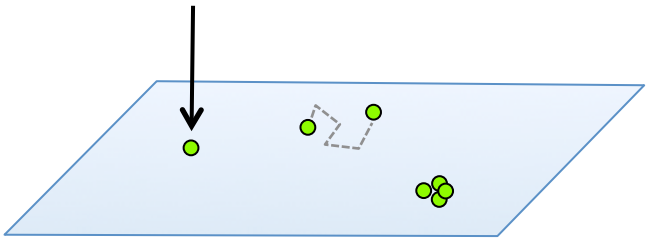
- GB gas retention due to bubble growth

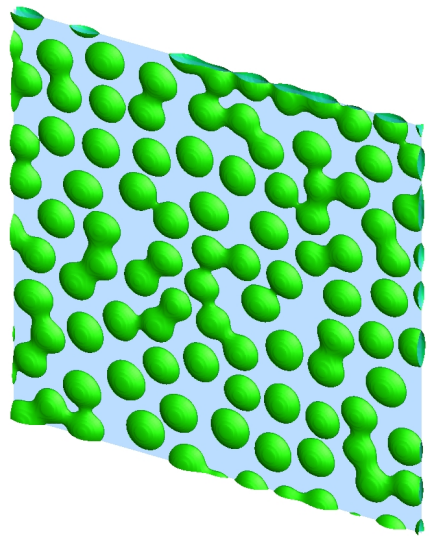
- Handled with saturation limit:

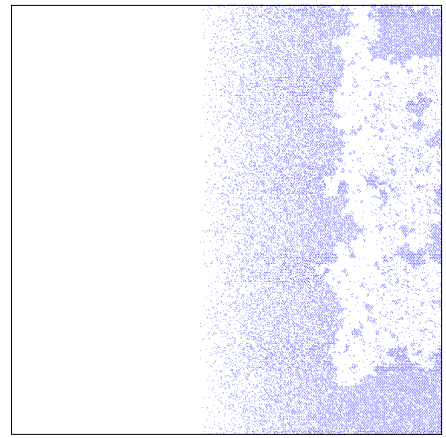
$$N^{sat} = \frac{4rf(\theta)X_{gb}^{c}}{3kTsin^2\theta}\left[\frac{2\gamma}{r} + P_{ext}\right]$$

Multi-scale Modeling

Simulation on 3 different scales







Nucleation

Random-Walk Monte Carlo

Growth & Coalescence

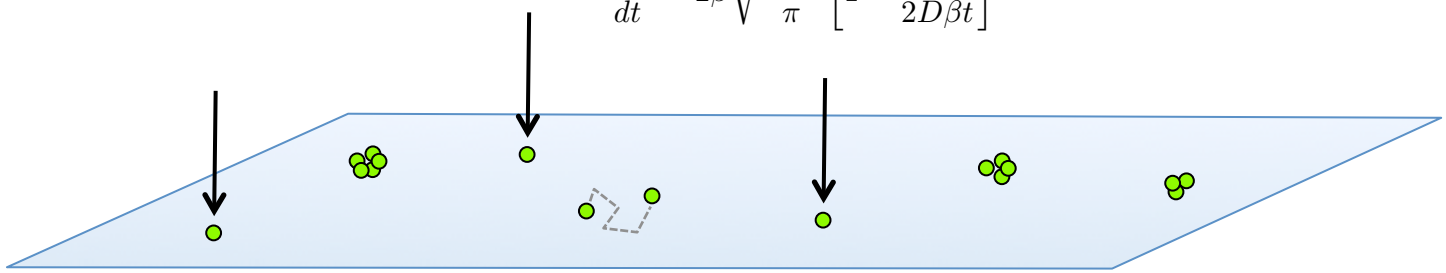
Phase-Field Method

Percolation on GB network

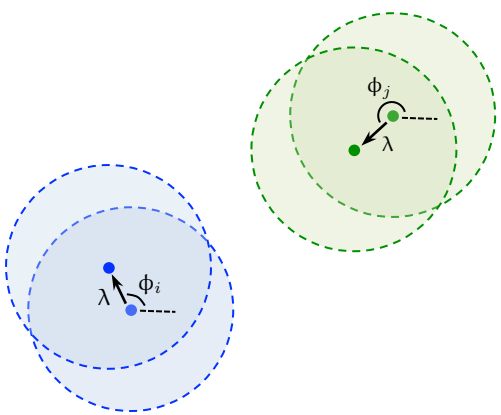
Percolation Model

Nucleation Model

$$\frac{dN}{dt} = 2\beta\sqrt{\frac{4Dt}{\pi}}\left[1 - \frac{b\delta N}{2D\beta t}\right]$$

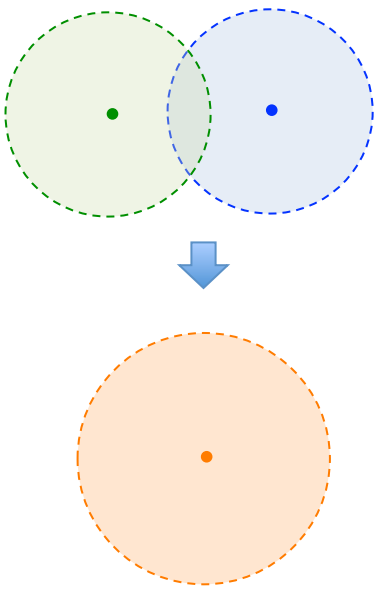


Random walk displacements



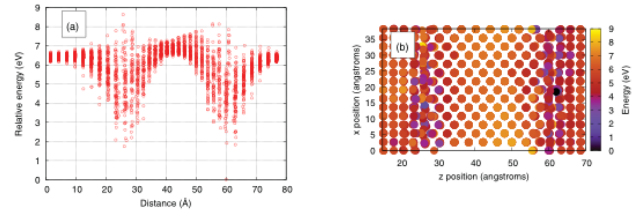
$$\lambda = \sqrt{4D_{gb}\Delta t}$$

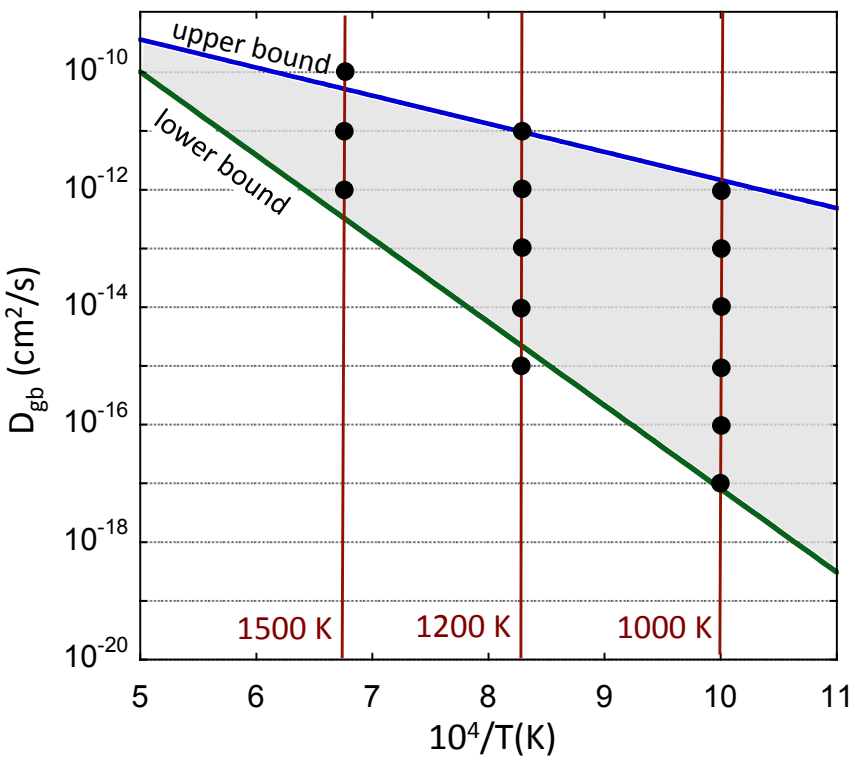
Coalescence

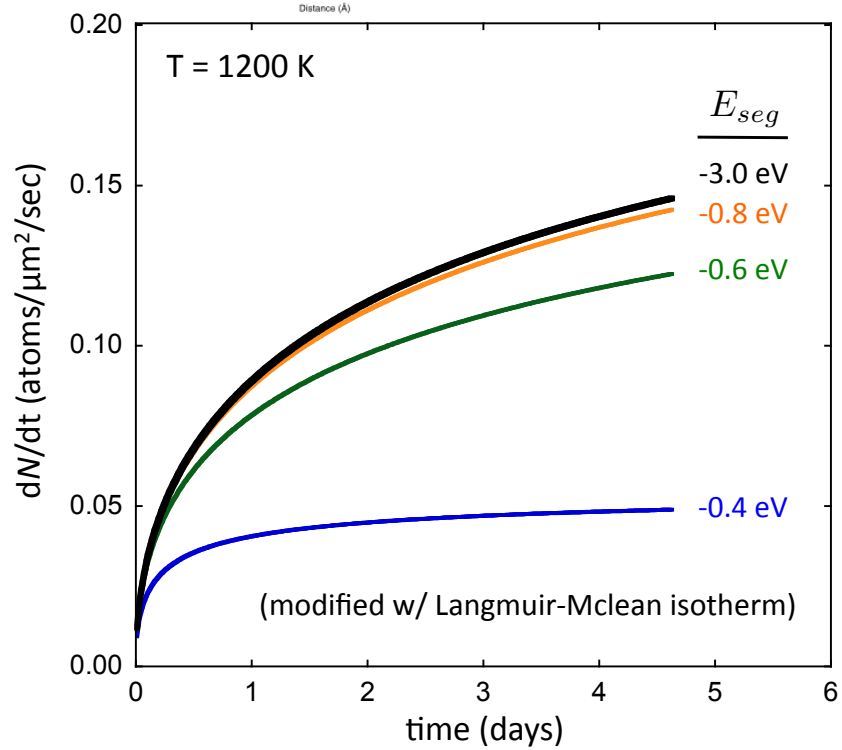


Nucleation Model

Nerikar et al. PRB 84, 174105 (2011)



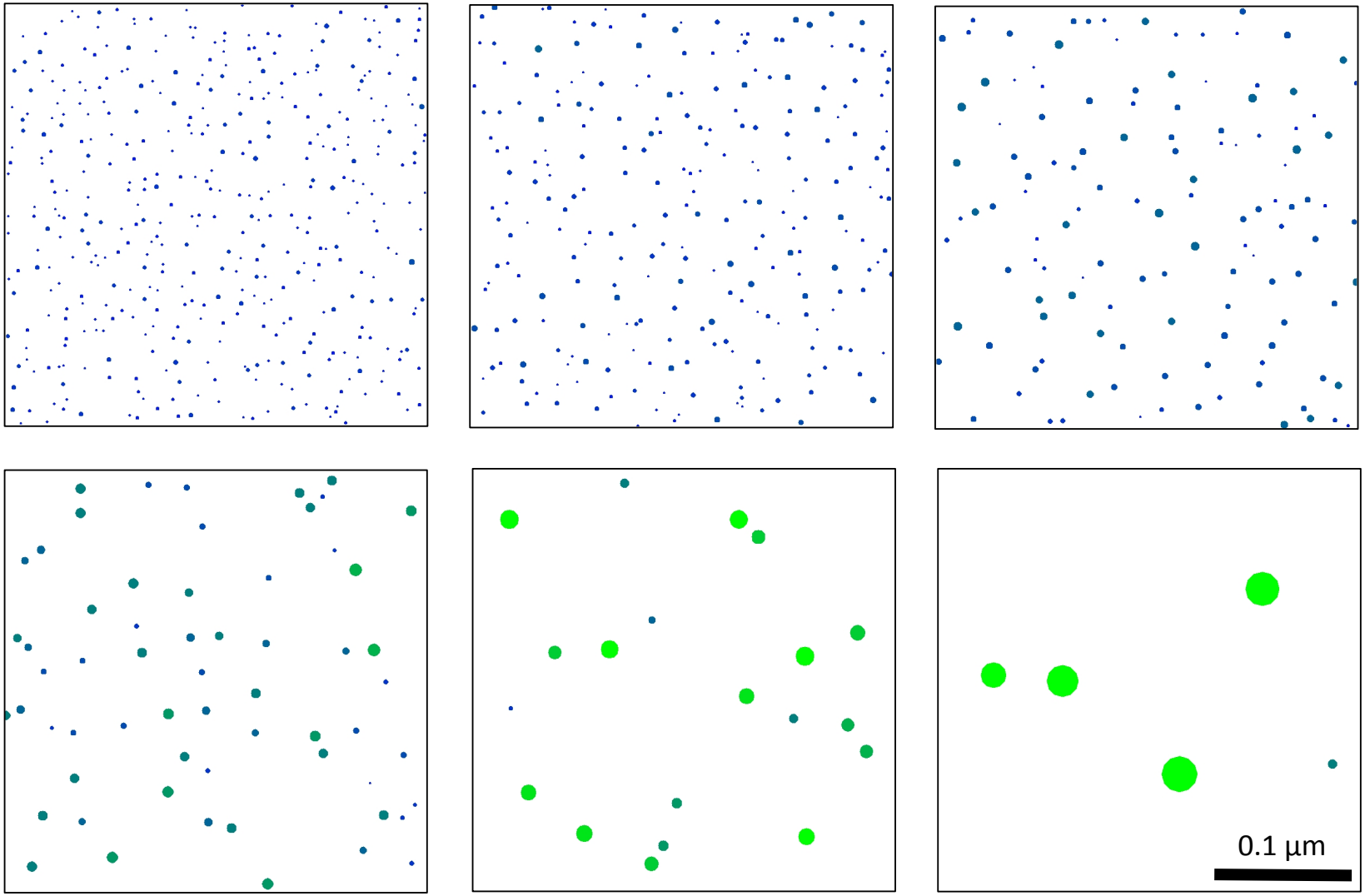




Three temperatures, wide range of D_{gb}

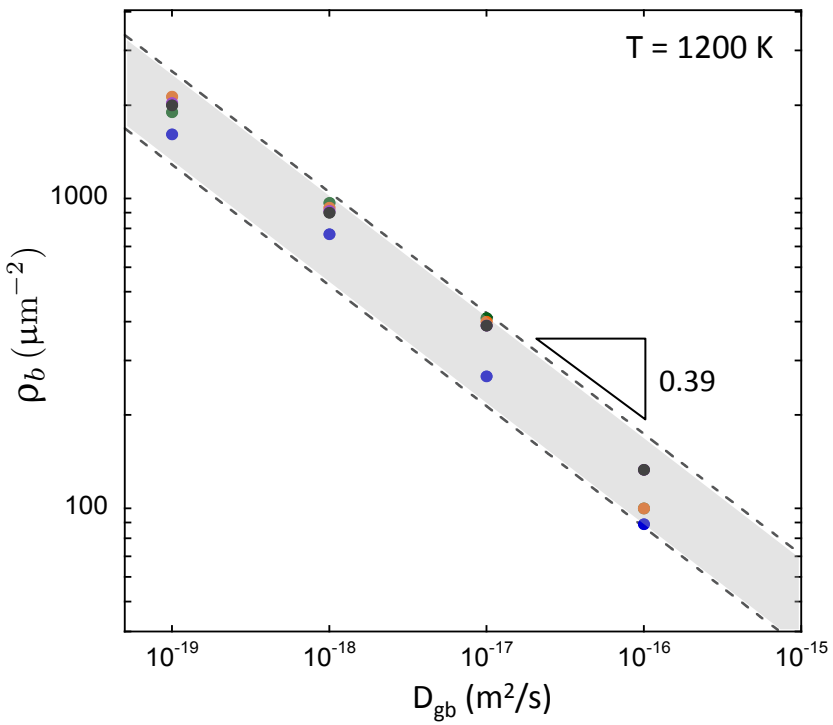
Flux varies with time and E_{seg}

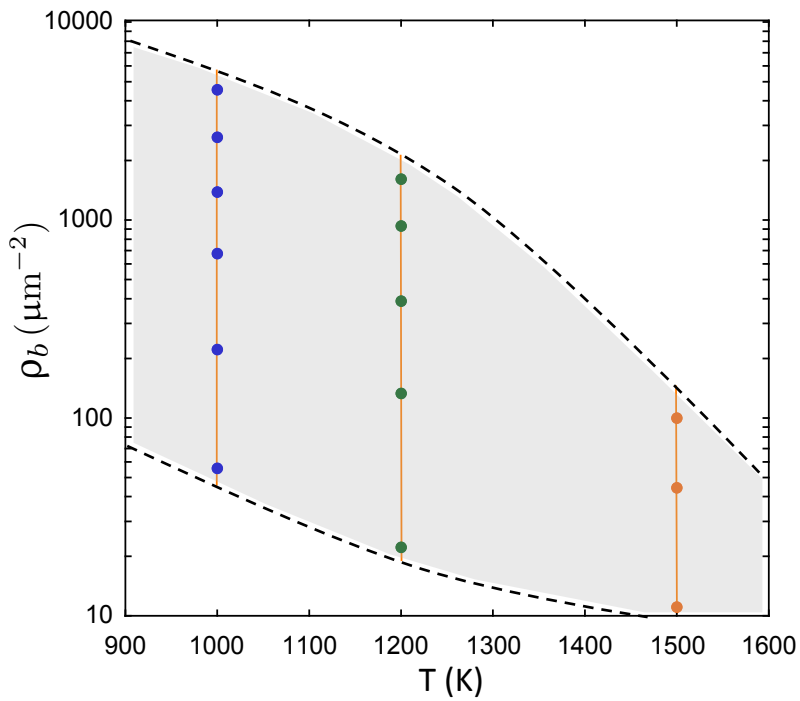
Nucleation Density



T = 1000 K; time = 2.3 days; coverage = 2%

Nucleation Density





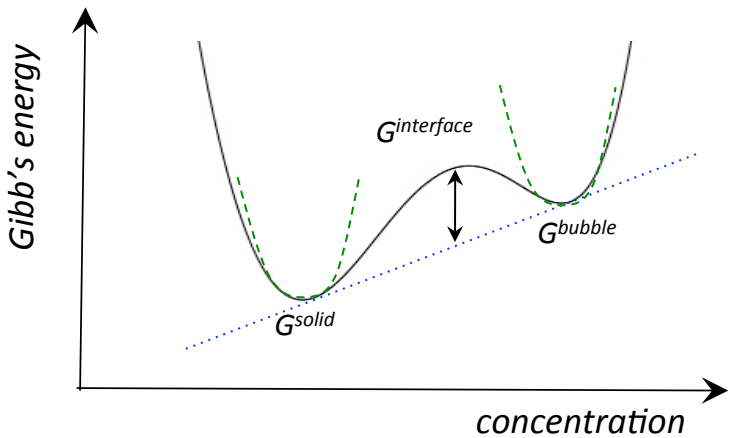
Bubble density varies mostly with D_{gb} (to a lesser extent with E_{seg})

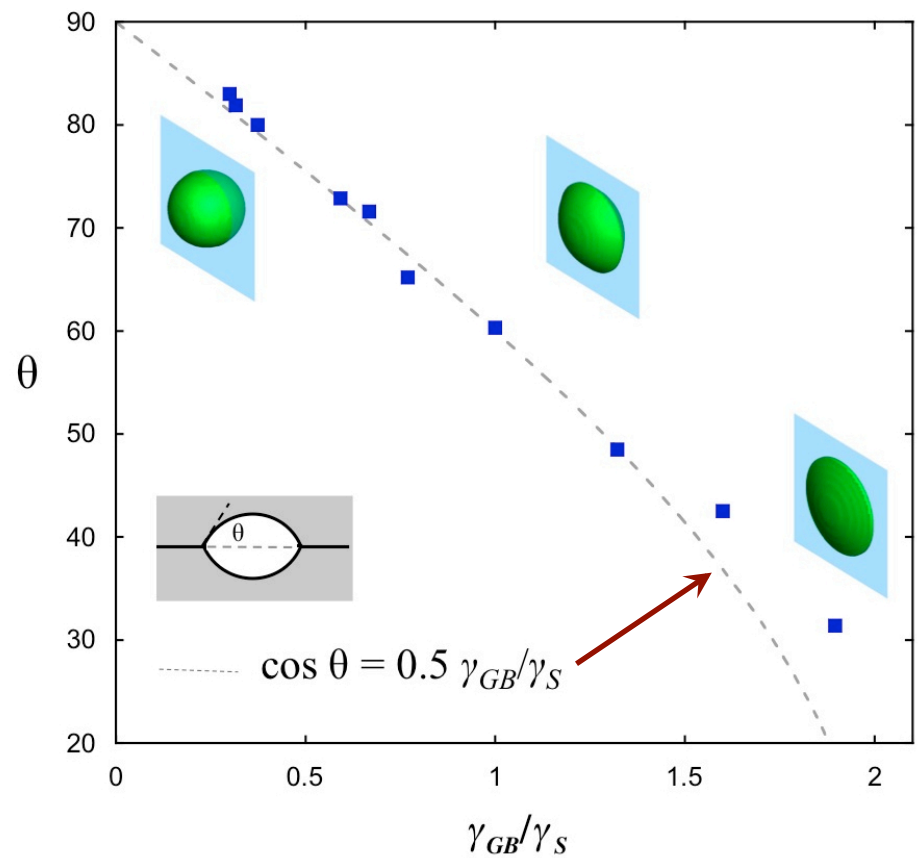
Increase Temp, decrease bubble density

Modeling Approach

Hypothesis: Grain boundary property variations are responsible for bubble percolation discrepancies.

Phase-Field Model

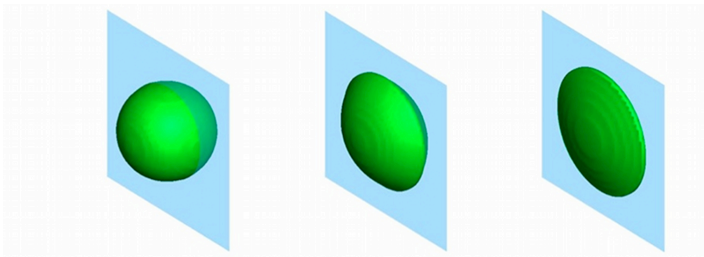




Parametric Study…

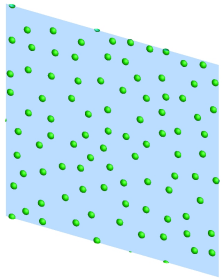
1.) CONTACT ANGLE

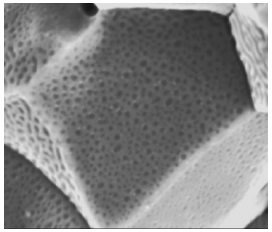
-> 40°, 50°, 60°, & 80°



2.) INITIAL BUBBLE DENSITY

-> 5, 10, 15, 20 μm^{-2} (agree with experiments)

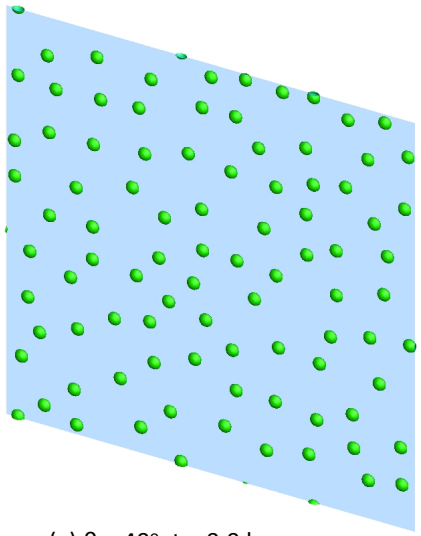




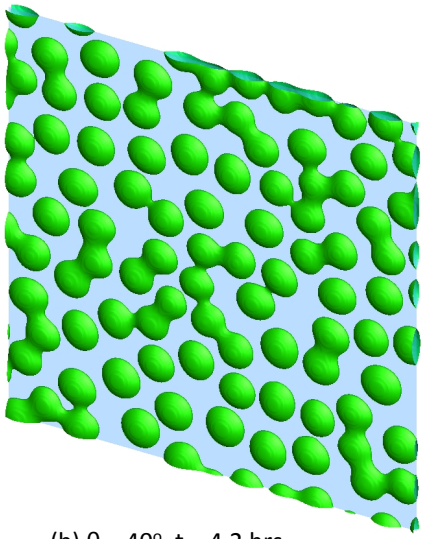
3.) GB GAS DIFFUSION

-> $D_{GB}/D_{lattice}$ = 0.1, 1.0, 5.0, 10.0, 15.0

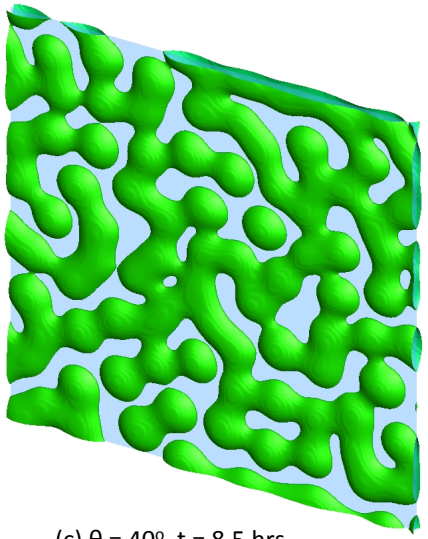
Bubble Percolation



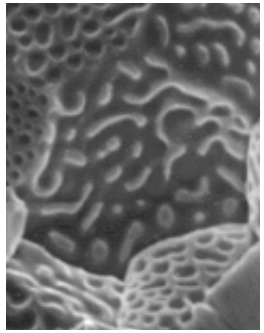
(a) θ = 40°, t = 0.0 hrs

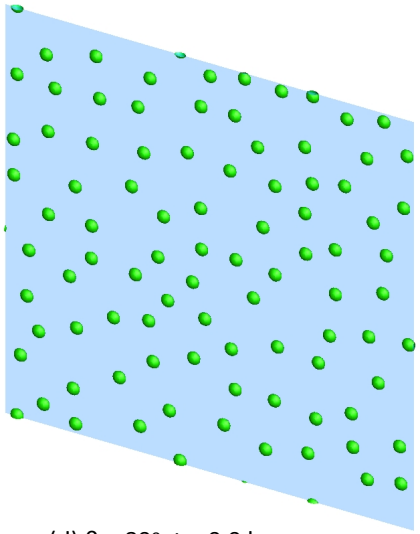


(b) θ = 40°, t = 4.2 hrs

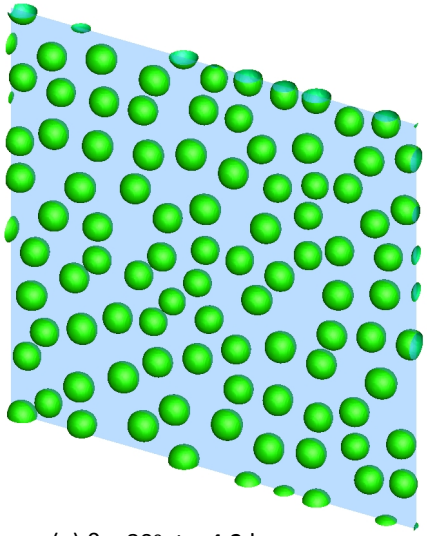


(c) θ = 40°, t = 8.5 hrs

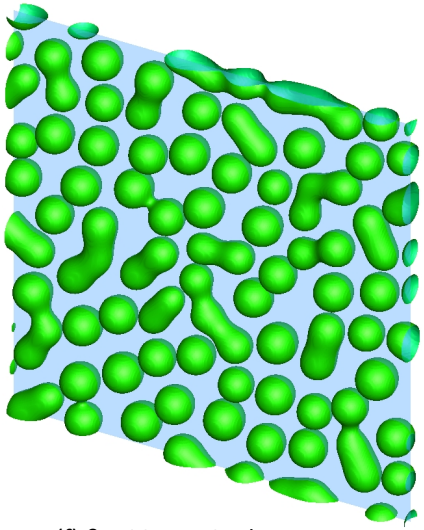




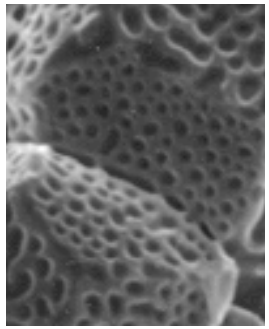
(d) θ = 80°, t = 0.0 hrs

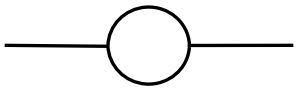


(e) θ = 80°, t = 4.2 hrs



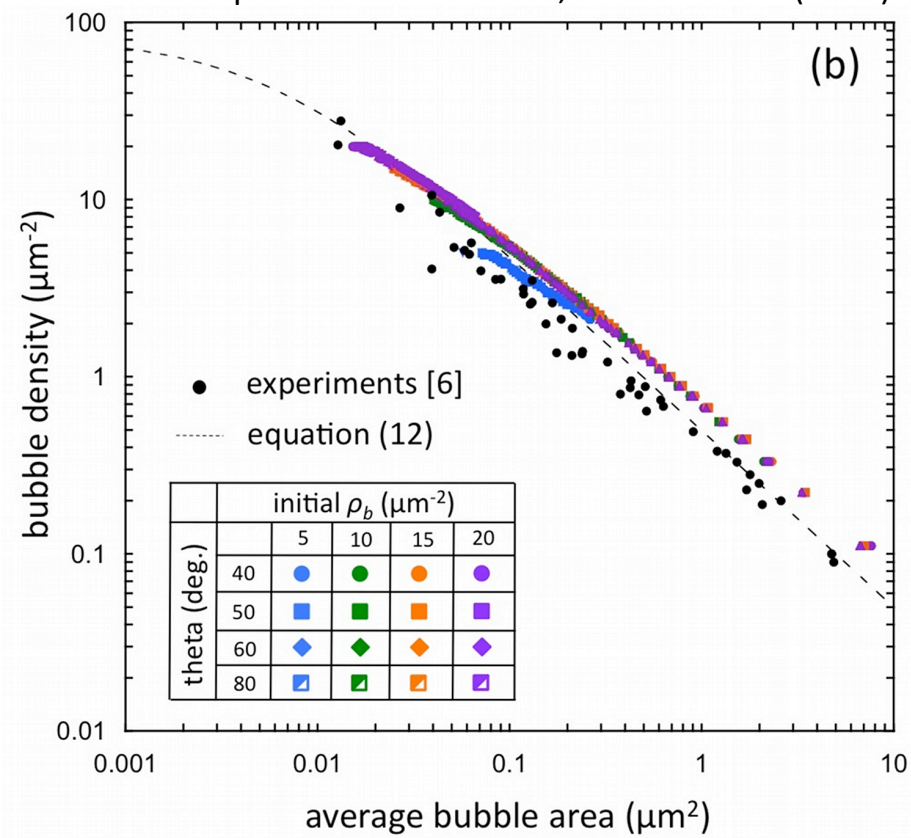
(f) θ = 80°, t = 8.5 hrs

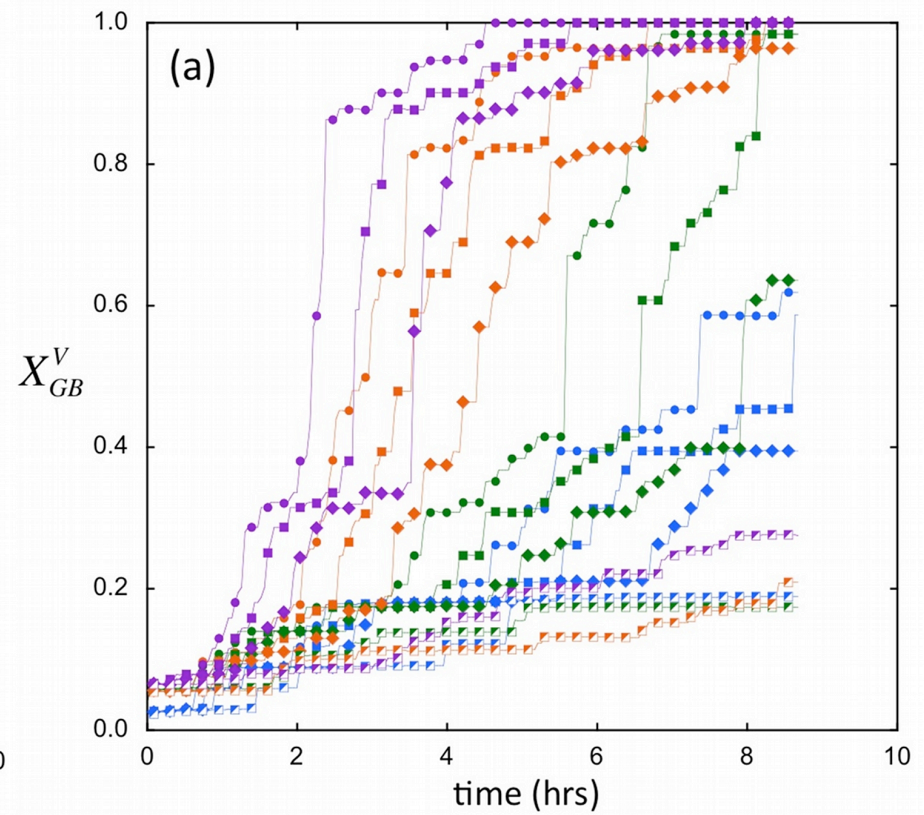




Bubble Percolation

Experiments: R.J. White, JNM 325: 133 (2004)

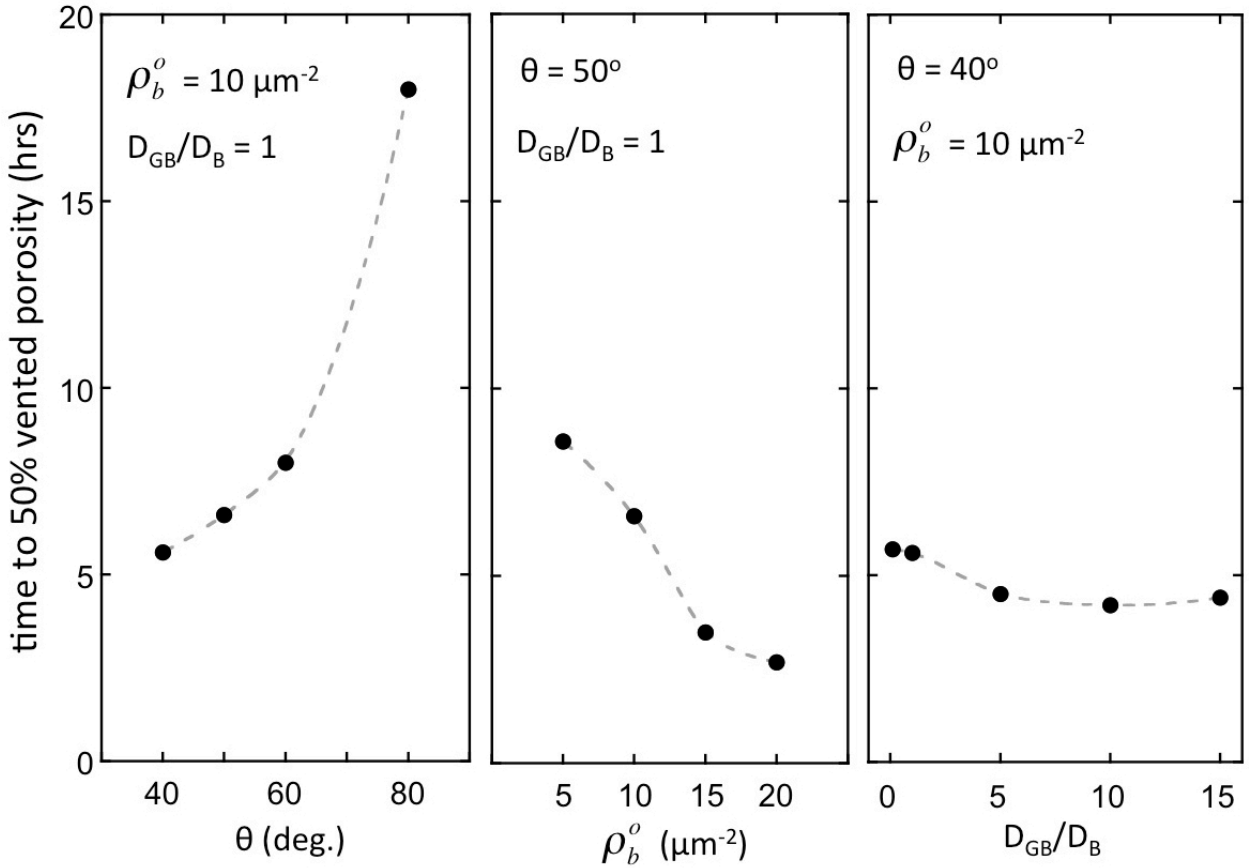




Agreement between simulation and experiments

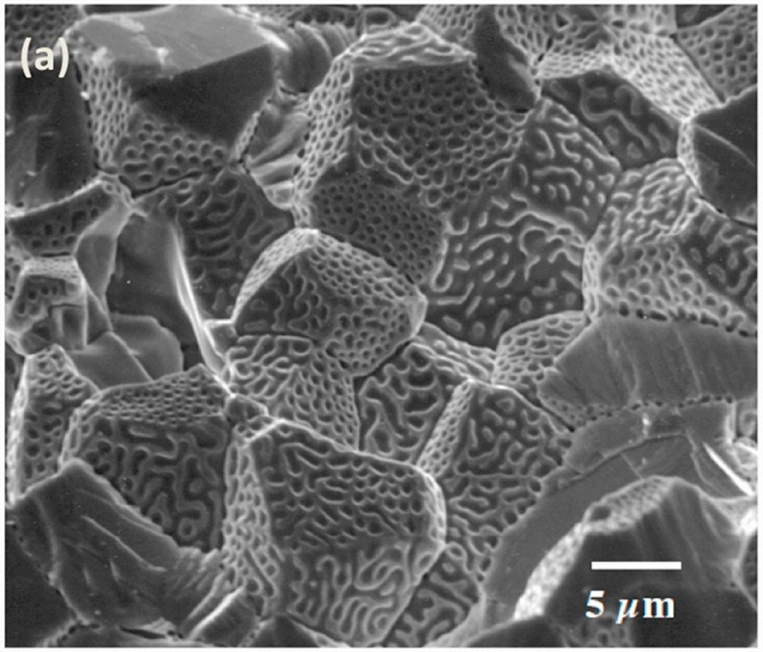
Large variability in the bubble percolation rate

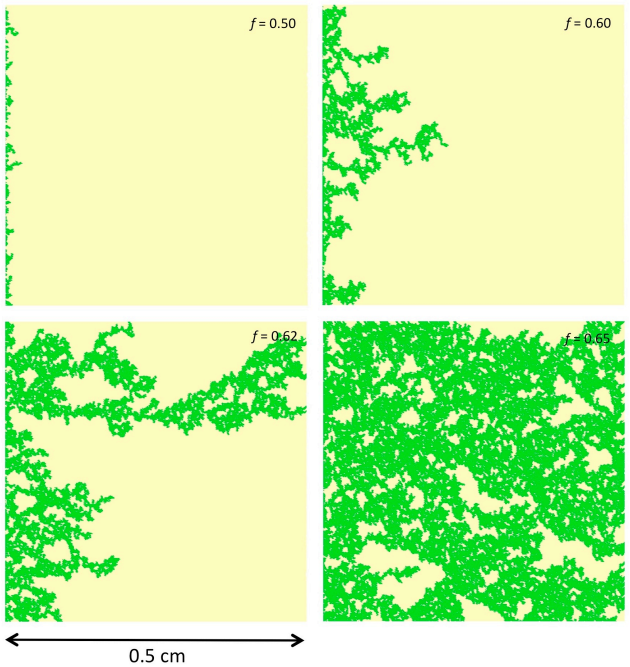
Bubble Percolation



Time to 50% vented porosity can vary by a factor of 3 with varying bubble shape and initial bubble density.

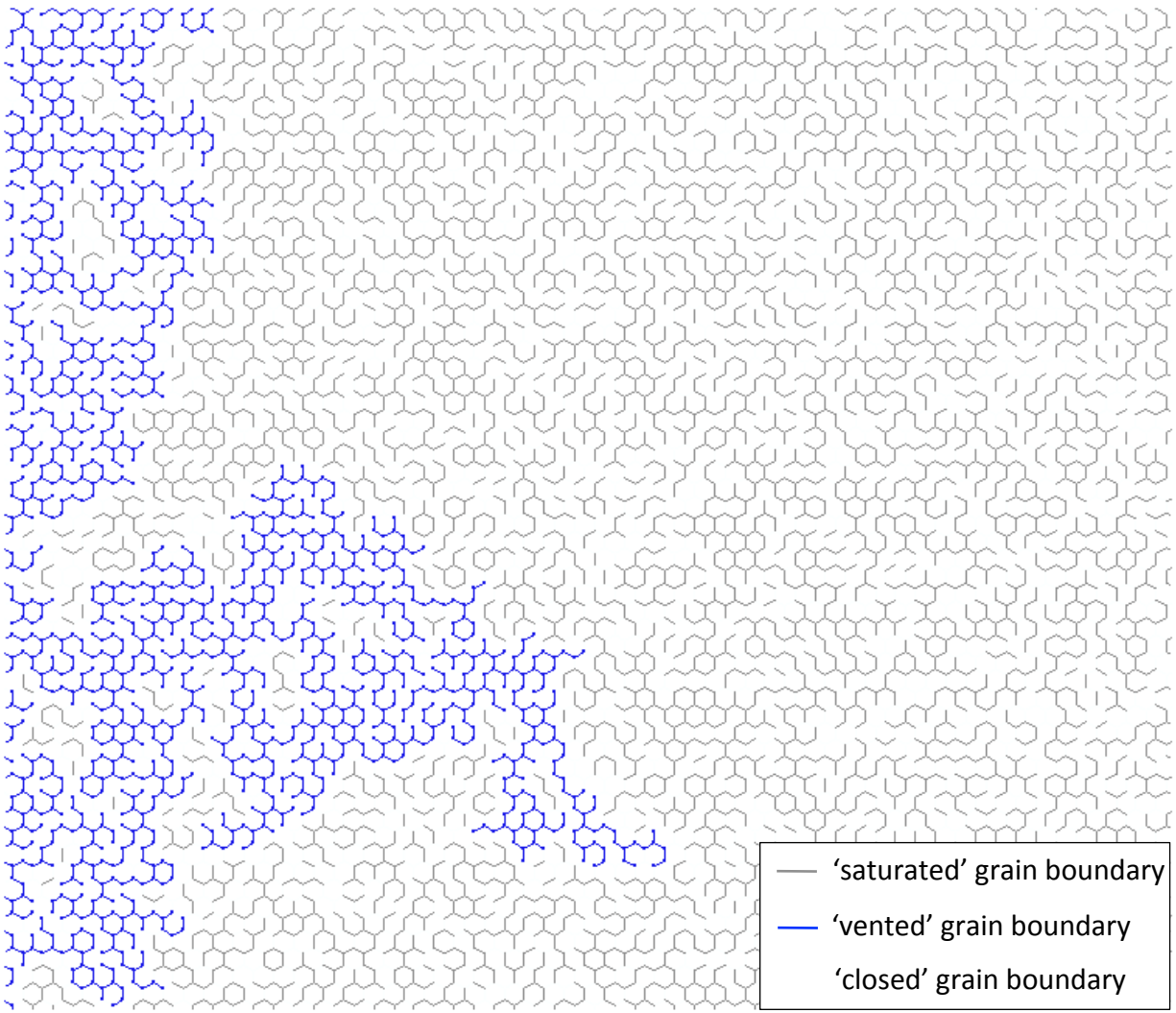
Longer-Range Fission Gas Transport

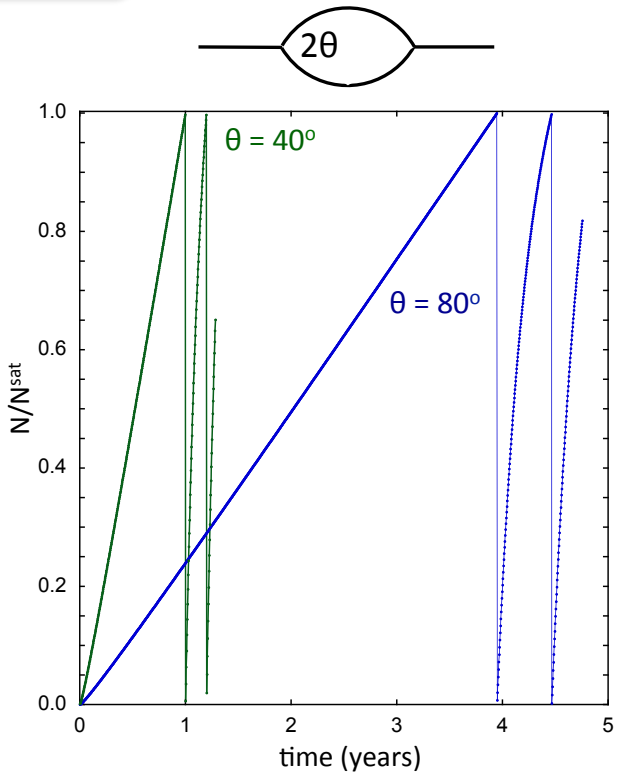




Percolation Model

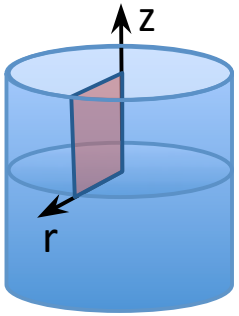
Incorporate a 3rd stage

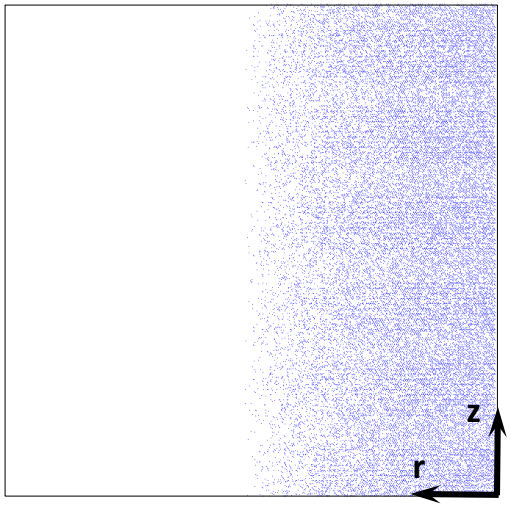




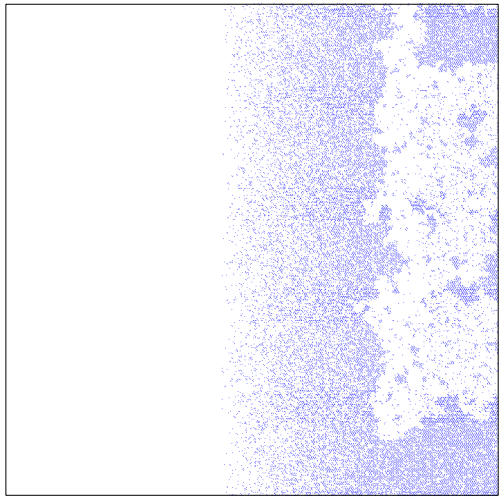
- Randomness in θ
- Search for pathways of 'vented' grain boundaries

Percolation Model

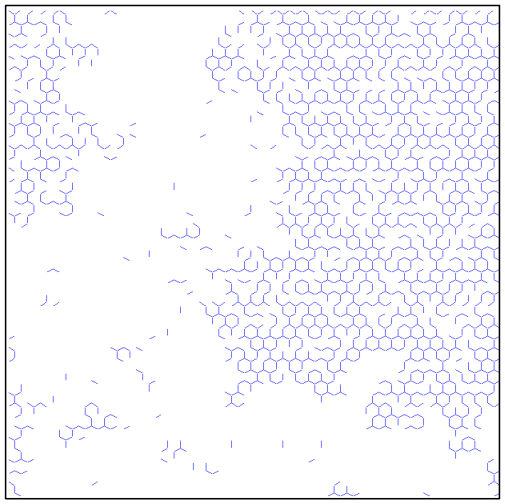




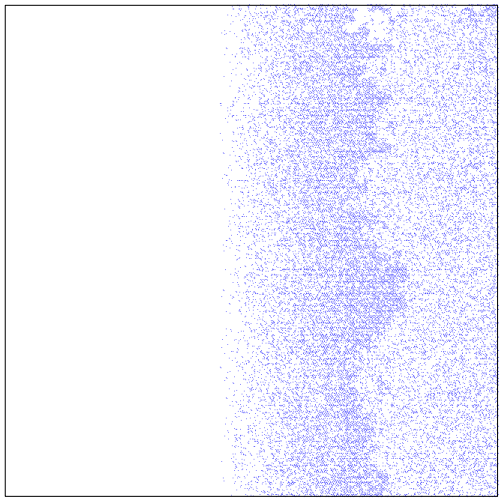
(a) t = 926 days



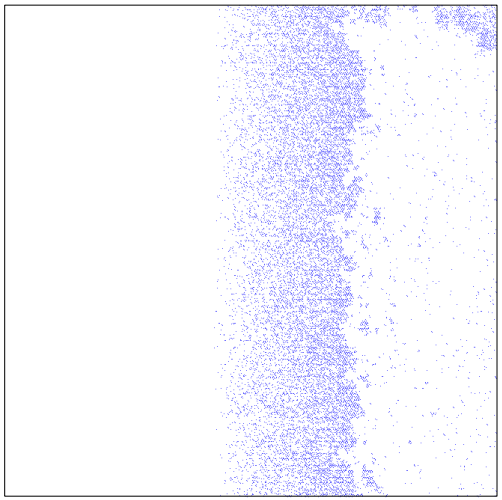
(b) t = 1157 days



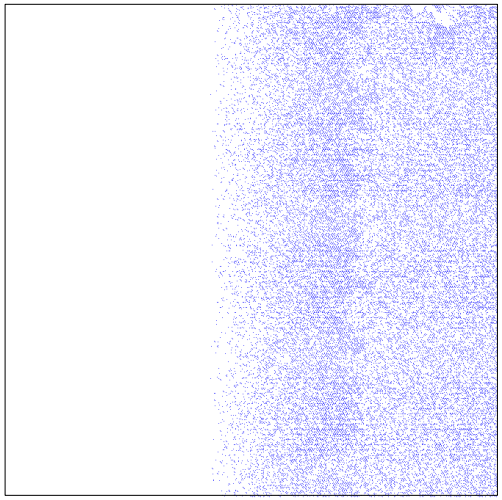
(c) t = 1157 days (close-up)



(d) t = 1208 days

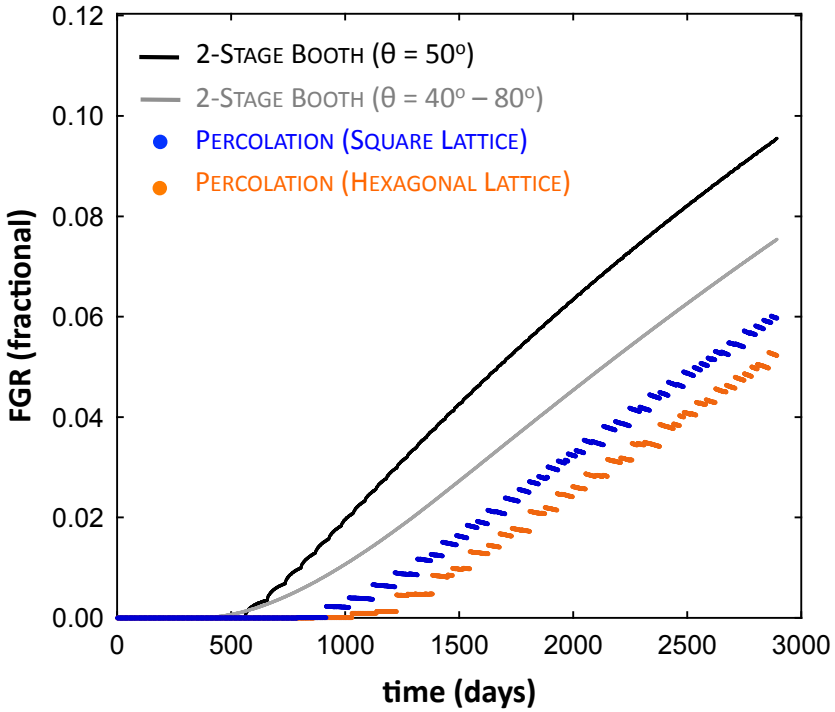


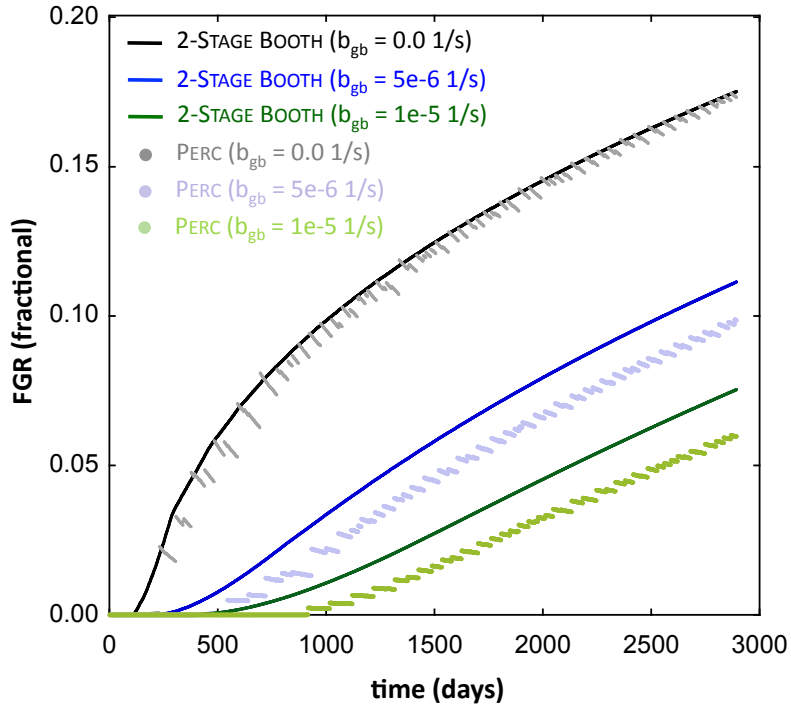
(e) t = 1250 days



(f) t = 1308 days

Fission Gas Release





Percolation reduces release rate

Reduction due to resolution effect

Thank you for your attention!