INL/MIS-20-57508-Revision-0

Phase-field simulation of intergranular fission gas bubble growth in U3Si2

March 2020

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Idaho National Laboratory

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Phase-field simulation of intergranular fission gas bubble growth in U_3Si_2

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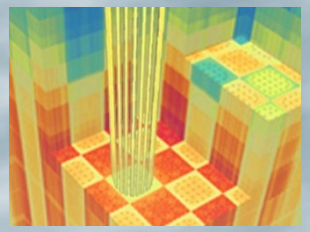
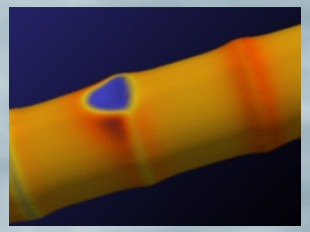
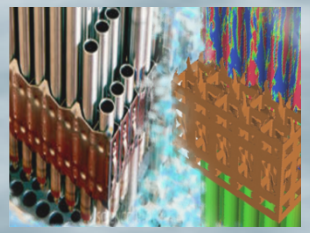




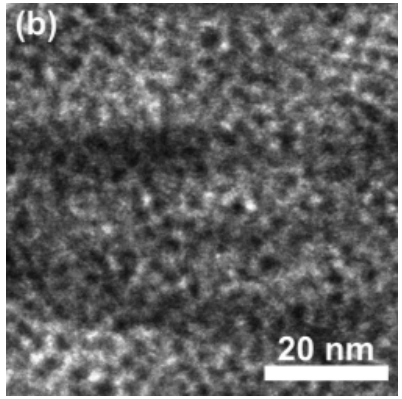

CASL The Consortium for Advanced Simulation of LWRs
A DOE Energy Innovation Hub

U.S. DEPARTMENT OF ENERGY

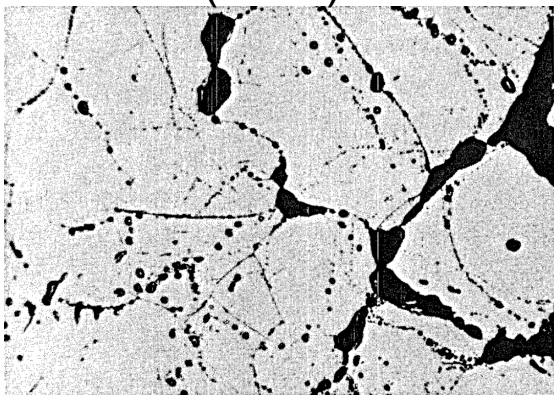
U_3Si_2 is being considered as a potential accident-tolerant fuel

- Compared with UO_2:
  - Lower melting temperature
  - But higher thermal conductivity may give higher margin to melting temperature
- U_3Si_2 swelling/fission gas release behavior less well characterized
  - Evidence from higher-temperature irradiation suggests pellet-form fuel would remain crystalline, have similar microstructure to UO_2 fuel
- BISON model recently developed based on these assumptions

Barani et al., J. Nuclear Mater., 522, 97-110 (2019)

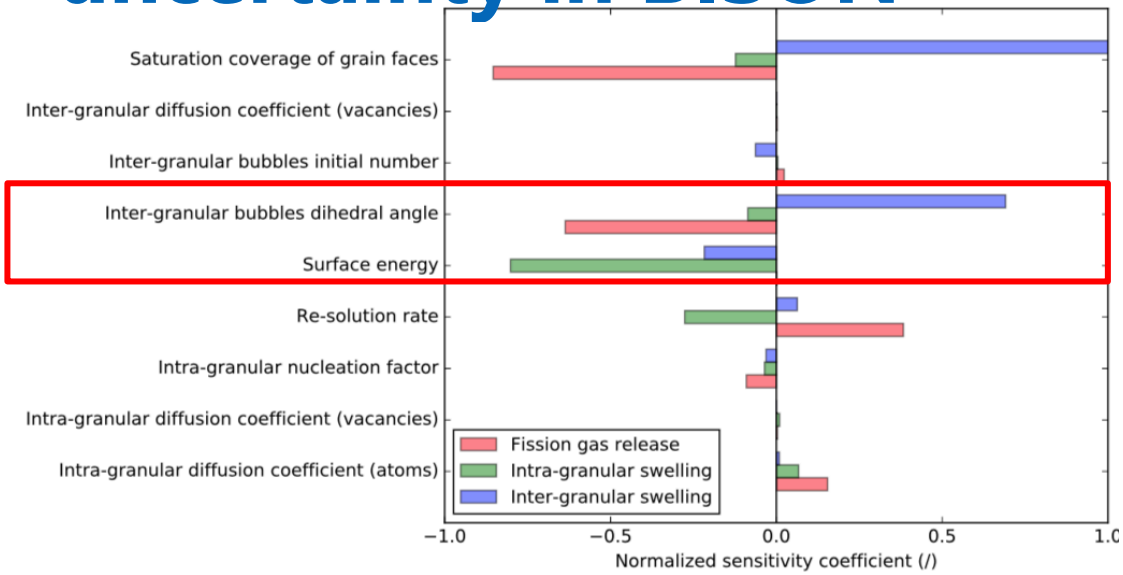


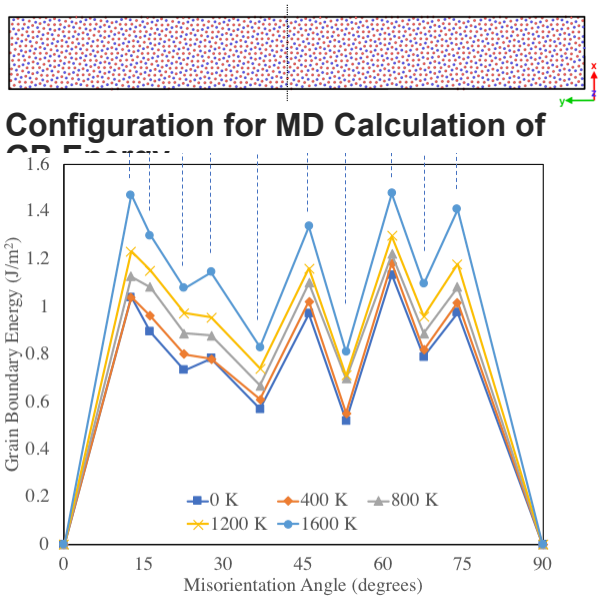
U_3Si_2 implanted with Xe at 873K (Miao et al., J. Nuclear Mater., 503, 314-322 (2018)



U_3Si_2 irradiated at ~950 K and ~6 GWd/tU (Shimizu, NAA-SR-1062, 1965)

Lower length scale calculations to reduce uncertainty in BISON

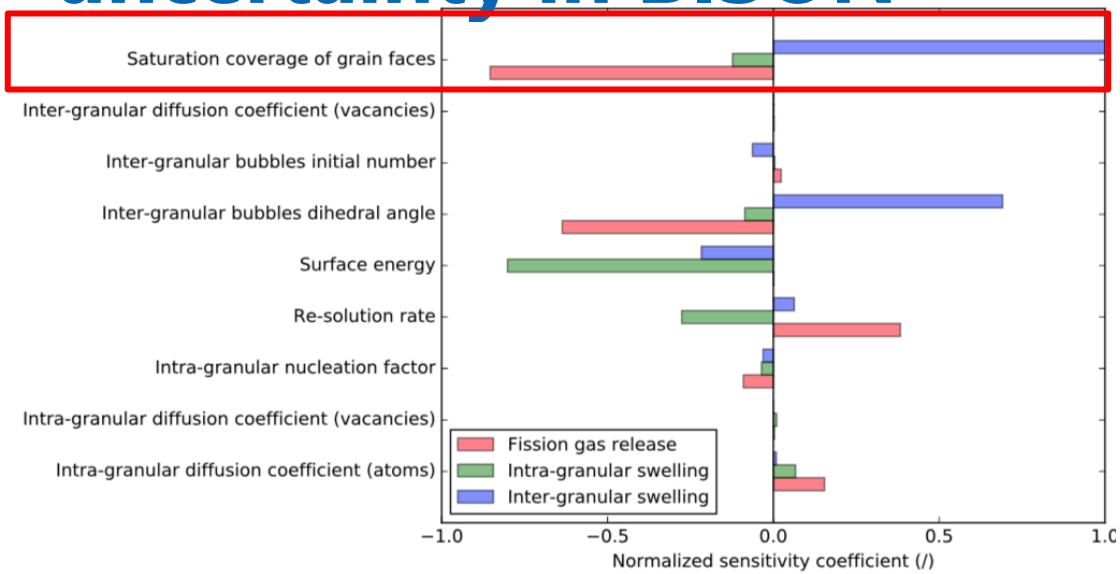


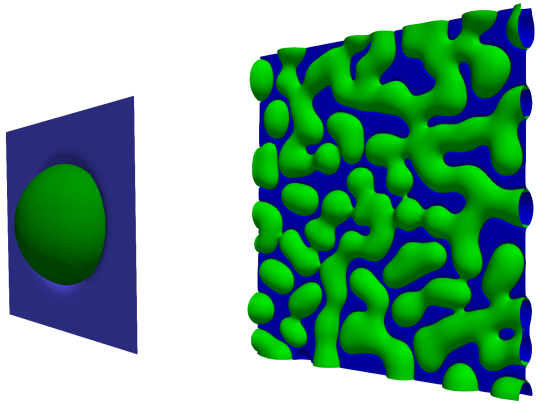


- Sensitivity analysis of Bison U_3Si_2 swelling and gas release predictions showed strong dependence on inter-granular bubble dihedral angle and surface energy
  - Measured values also not available
- Surface energy and grain boundary energies were determined for U_3Si_2 using molecular dynamics (MD) calculations
  - Dihedral angle (θ) calculated from surface energy and grain boundary energy; input to Bison
  - Data also used to parameterize the Marmot phase-field model

Beeler, Baskes, Andersson, Cooper, Y. Zhang, *J. Nucl. Mater.*, 514, 290-298 (2019)

Lower length scale calculations to reduce uncertainty in BISON





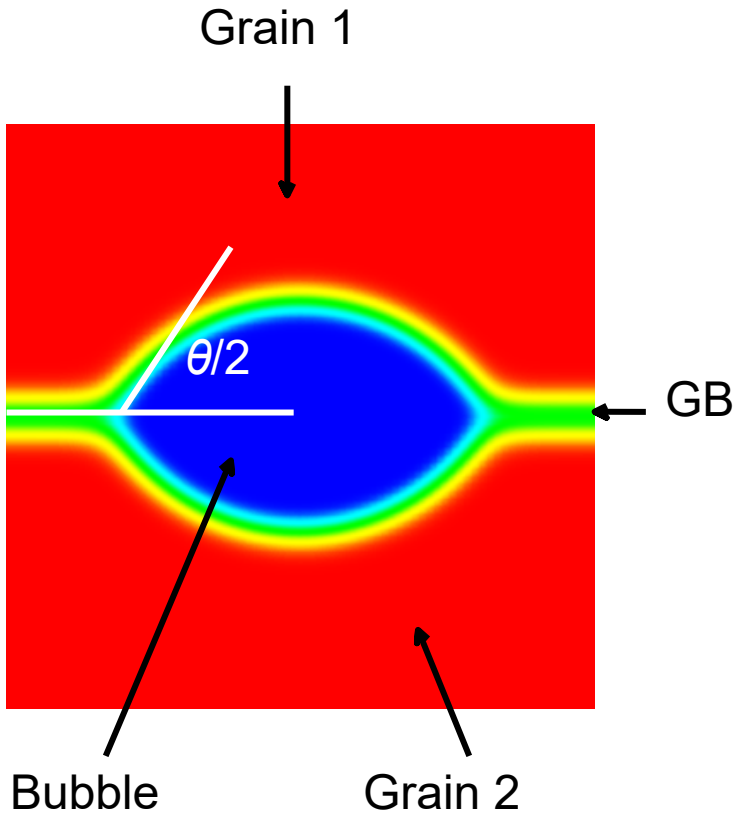
MARMOT

- Sensitivity analysis also showed strong dependence on saturation coverage of grain faces ($F_{c,sat}$)
  - No measured value available for U_3Si_2
- Phase-field simulations[1] showed progress of grain boundary venting was strongly dependent on intergranular bubble areal density and dihedral angle
  - New phase-field simulations are being used to determine $F_{c,sat}$ using U_3Si_2 parameters

[1]Millett, Tonks, Biner, L. Zhang, Chockalingham, Y. Zhang, *J. Nucl. Mater.*, 425, 130-135 (2012)

Phase-Field Model: Essential Physics

- Represent bubble phase and multiple grains of U_3Si_2
- Track vacancies and fission product species (Xe only)
  - Source terms for production
- Set surface energy and grain boundary energy
  - Controls dihedral angle θ
  - Remove bulk energy contribution to interfacial energy



Phase-Field Model: Grand-Potential Functional

$$\Omega = \int_V \left(m \left[\sum_\alpha \sum_{i=1}^{p_\alpha} \left(\frac{\eta_{\alpha i}^4}{4} - \frac{\eta_{\alpha i}^2}{2} \right) + \sum_\alpha \sum_{i=1}^{p_\alpha} \left(\sum_\beta \sum_{j=1, \alpha i \neq \beta j}^{p_\beta} \frac{\gamma_{\alpha i \beta j}}{2} \eta_{\alpha i}^2 \eta_{\beta j}^2 \right) + \frac{1}{4} \right] + \frac{\kappa}{2} \sum_\alpha \sum_{i=1}^{p_\alpha} |\nabla \eta_{\alpha i}|^2 + \sum_\alpha h_\alpha \omega_\alpha \right) dV$$

- Multi-phase, multi-order parameter extension to grand-potential model
- Advantages:
  - Bulk free energy contribution is removed from interfacial energy
  - Allows interfacial thickness and energy to be set independently, enabling coarser mesh, improved computational performance
  - Similar to KKS in this respect, but do not need separate phase concentration variables, so performance is improved
  - Prevents spurious formation of additional phases at two-phase interfaces

L.K. Aagesen, Y. Gao, D. Schwen, K. Ahmed, *Phys. Rev. E*, 98, 023309 (2018).
L.K. Aagesen, D. Schwen, M.R. Tonks, Y. Zhang, *Comp. Mat. Sci.*, 161, 35-45 (2019).

Phase-Field Model Evolution Equations

- Order parameters: Allen-Cahn
- Densities: change to chemical potential for each species

Gas:

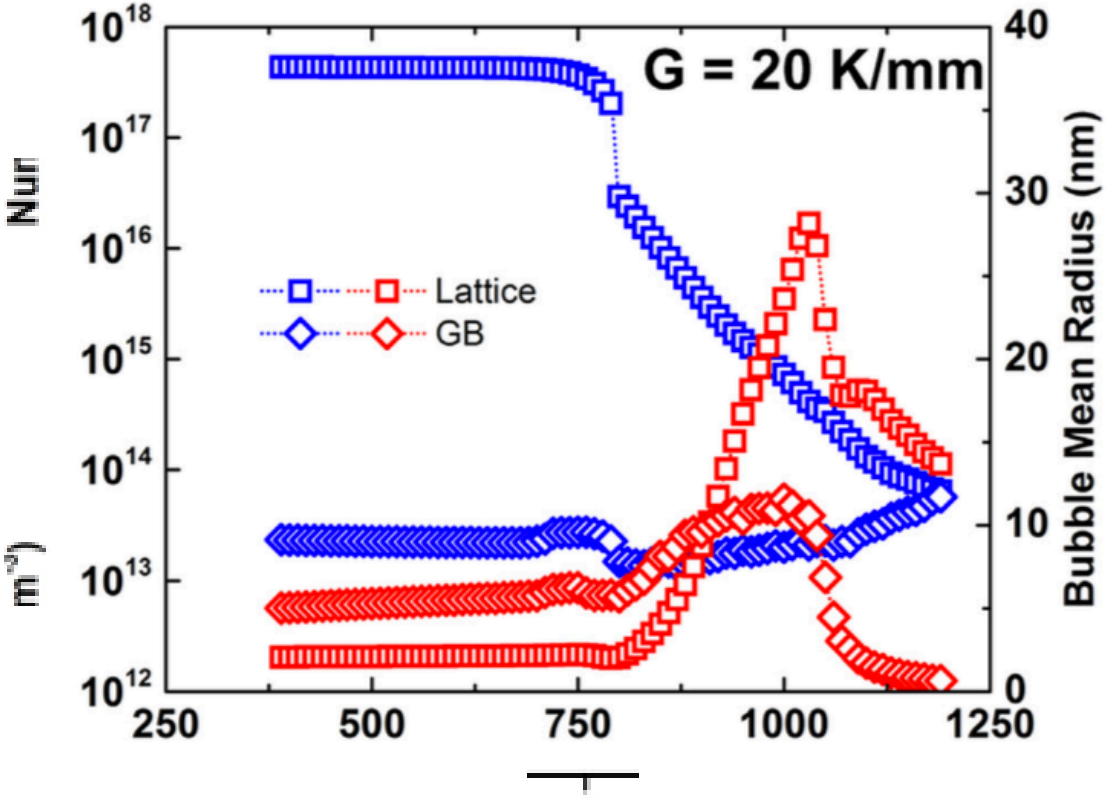
$$\frac{\partial \mu_g}{\partial t} = \frac{1}{\chi_g}\left[\nabla \cdot (D_g \chi_g \nabla \mu_g) + s_g - \sum_{\alpha}\sum_{i=1}^{p_\alpha} \frac{\partial \rho_g}{\partial \eta_{\alpha i}} \frac{\partial \eta_{\alpha i}}{\partial t}\right]$$

Vacancies:

$$\frac{\partial \mu_v}{\partial t} = \frac{1}{\chi_v}\left[\nabla \cdot (D_v \chi_v \nabla \mu_v) + s_v - \sum_{\alpha}\sum_{i=1}^{p_\alpha} \frac{\partial \rho_v}{\partial \eta_{\alpha i}} \frac{\partial \eta_{\alpha i}}{\partial t}\right]$$

Phase-Field Model Initial Conditions

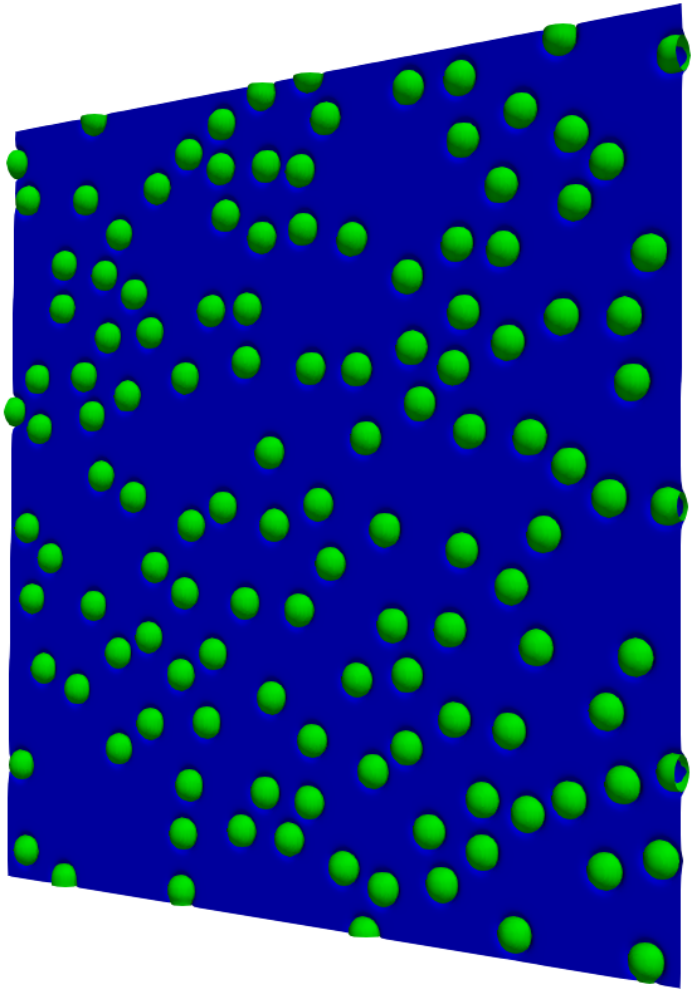
- Intergranular bubble areal density (n_a): determine from rate theory simulations
  - At 1035 K, n_a = 15 / μm^2



Y. Miao, K.A. Gamble, D. Andersson, B. Ye, Z.-G. Mei, G. Hoffman, A.M. Yacout, *Nucl. Eng. Design,* 322, 336-344 (2017).

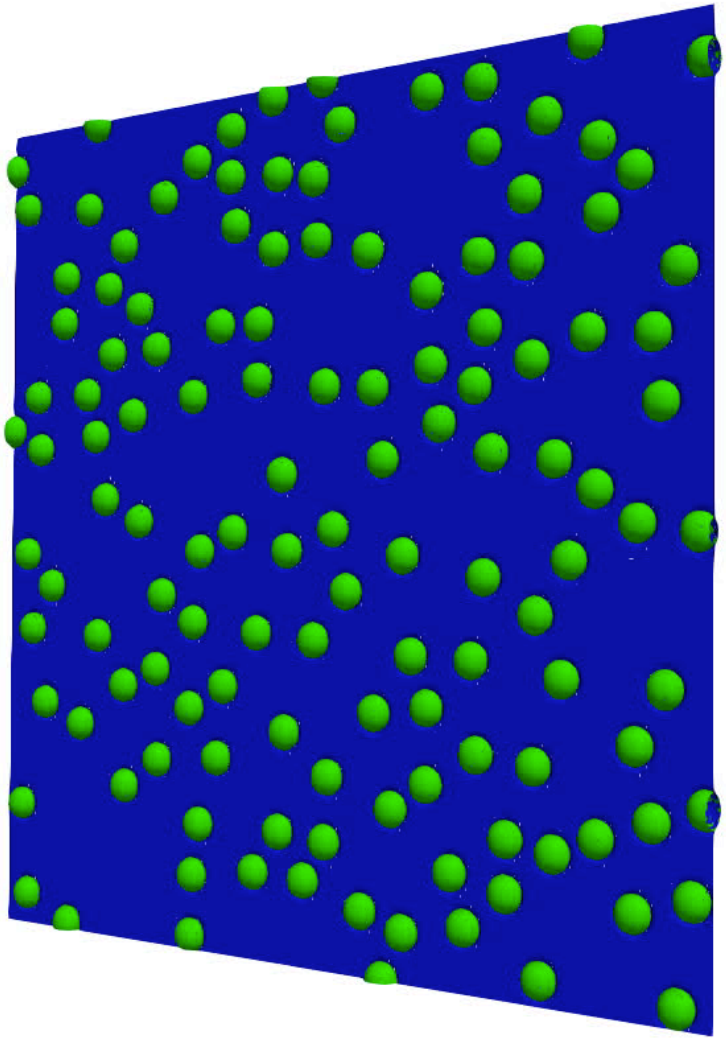
Phase-Field Model Initial Conditions

- Determine $F_{c,sat}$
- 1035 K
- $\theta/2 = 73$
- No-flux Boundary Conditions
- 3 μm × 3 μm grain boundary
- Populate with randomly placed lenticular bubbles, n_a = 15 / μm^2, minimum spacing 160 nm



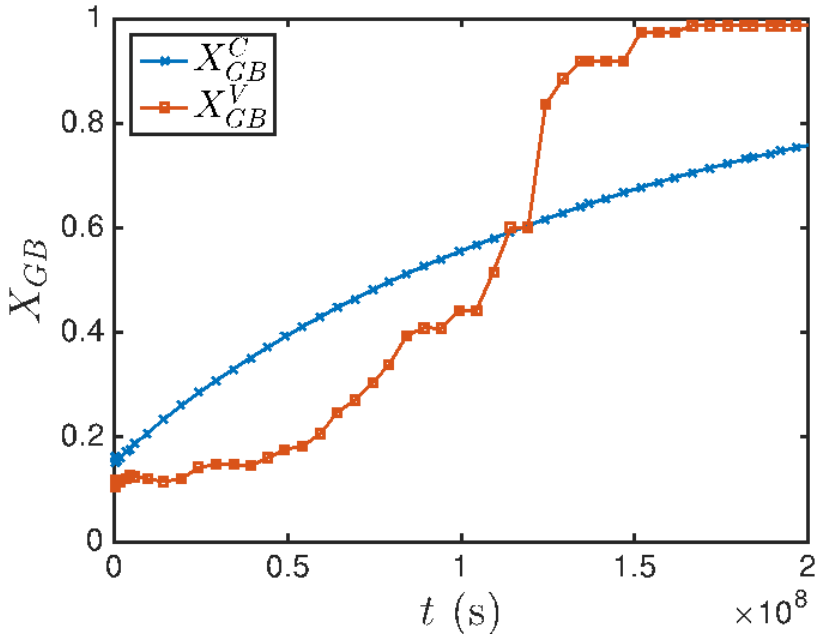
3 μm × 3 μm

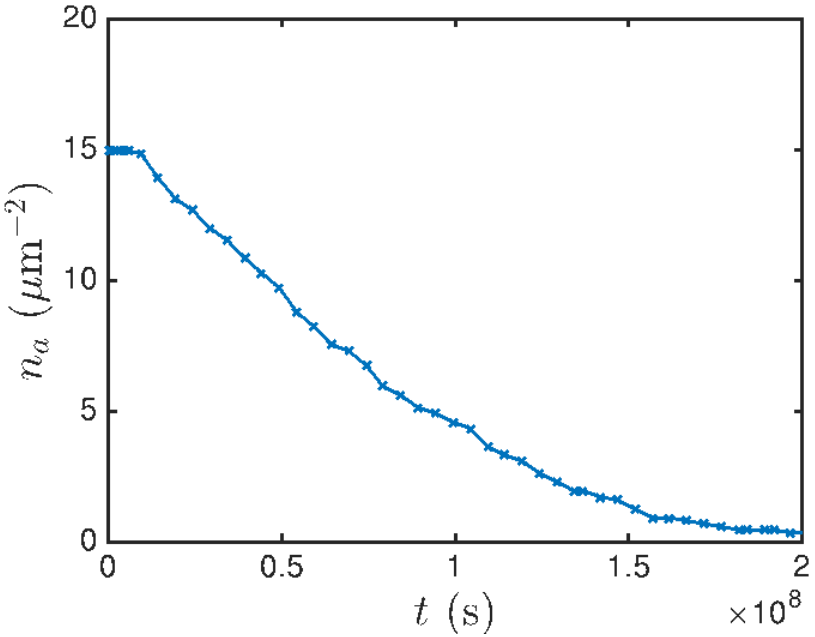
Phase-Field Simulation Results



3 μm × 3 μm

Phase-Field Simulation Results

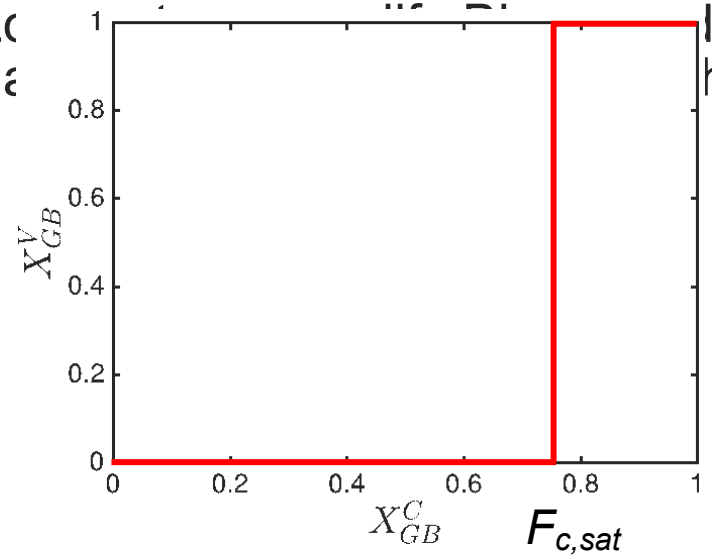




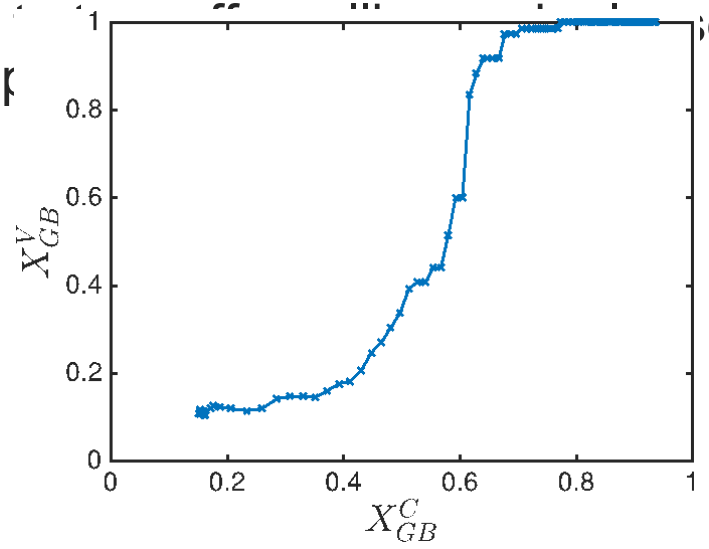
- Plot fractional coverage of GB (X_{GB}^{C}) and fraction of bubbles that are vented to edge of domain (X_{GB}^{V}) vs. time
  - Less rapid increase with respect to time compare to previous simulations of Millett et al. due to to slow buildup from source terms
- Areal density of bubbles versus time
  - Rate of coalescence relatively constant until the bubble density reaches approximately half its initial value, then slows

Informing BISON with Phase-Field Results

- Plot fraction of bubbles that are vented to edge of domain (X_{GB}^{V}) versus fractional coverage of GB (X_{GB}^{C})
- Implications for Bison:
  - Short term: set $F_{c,sat}$ where slope of curve is greatest (shown: X_{GB}^{C} = 0.62)
  - Lo[illegible] ga[illegible]



Bison assumption



Marmot Simulation

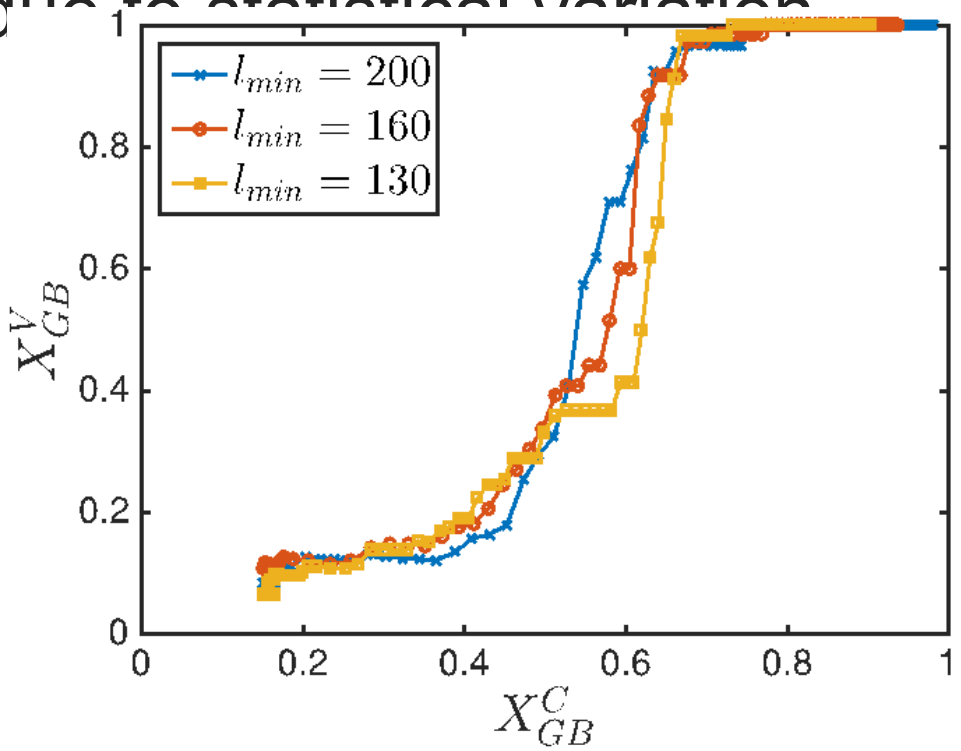
Effect of Simulation Assumptions on Predicted Value for BISON Model

- Simulation initial conditions
  - Maintain all simulation parameters the same including minimum spacing l_{min} = 160 nm
  - Change seed in random number generator used to determine initial bubble positions
  - 5 total configurations simulated using these parameters
- Mean $F_{c,sat}$ = 0.60
- Standard deviation indicates calculated value of $F_{c,sat}$ is relatively insensitive to initial bubble configuration

Configuration	$F_{c,sat}$
1	0.54
2	0.62
3	0.61
4	0.63
5	0.62
Mean	0.60
Standard Deviation	0.036

Effect of Minimum Bubble Spacing in Initial Conditions

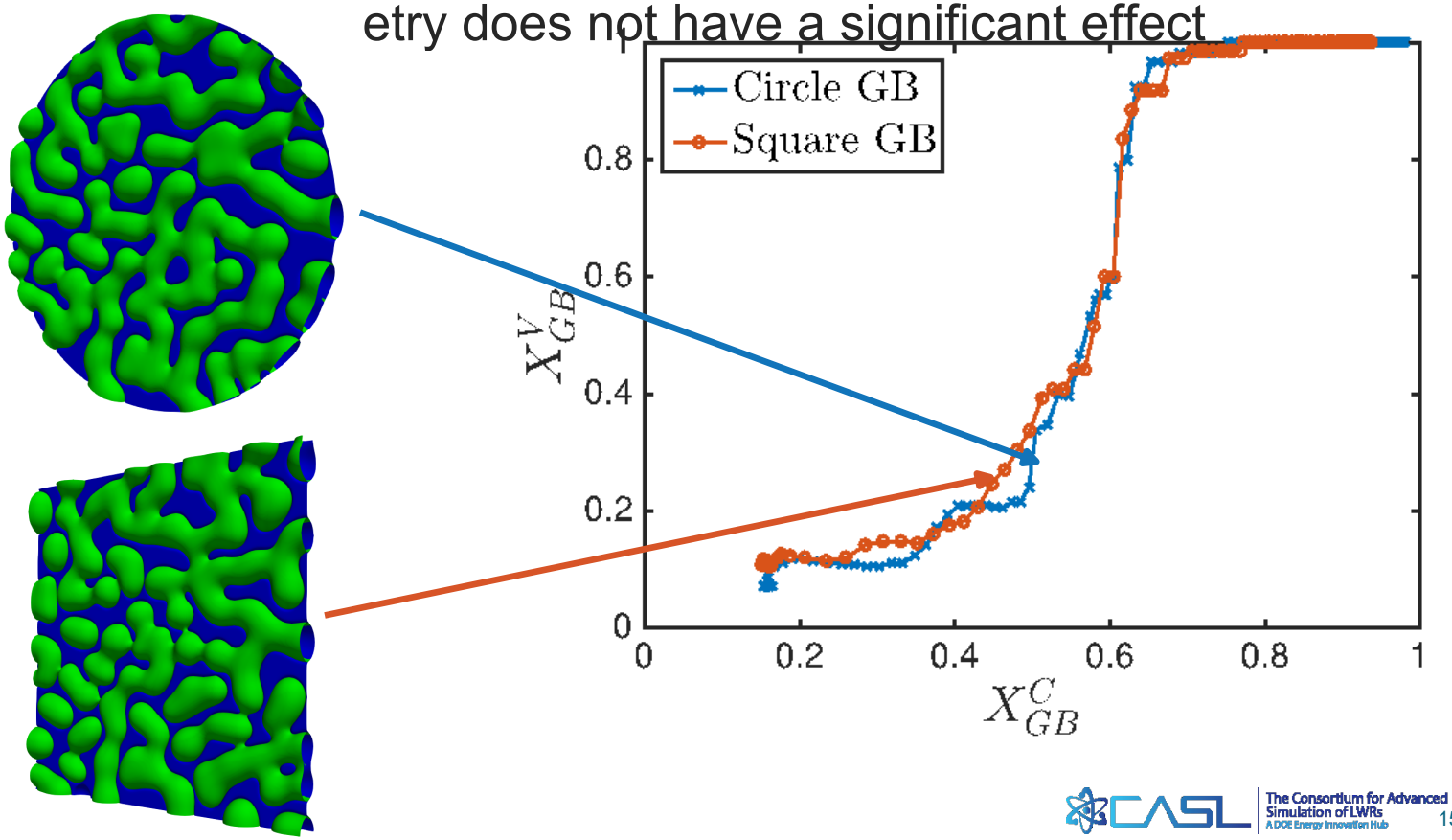
- Also simulated l_{min} = 130 nm, 200 nm, 5 configurations each
- 200 nm: Initial portion of release curve delayed
- Slight decrease in $F_{c,sat}$ with l_{min} , but may be just due to statistical variation



Min. spacing (l_{min}), nm	$F_{c,sat}$
130	0.61 ± 0.039
160	0.60 ± 0.036
200	0.58 ± 0.046

Effect of simulation domain geometry

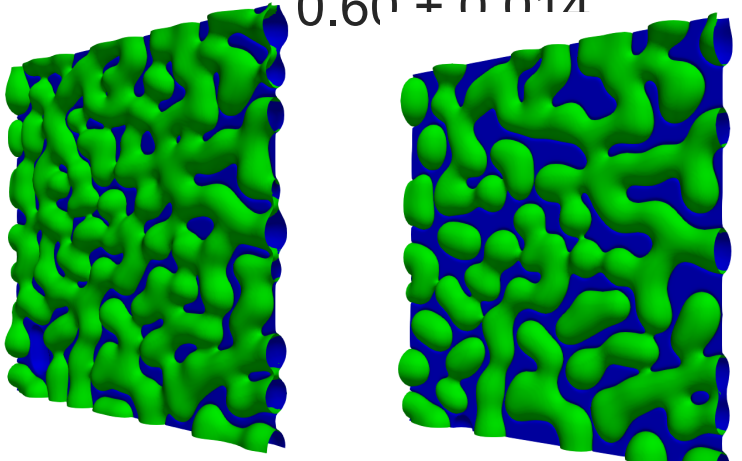
- Compare venting curves for circular GB versus square GB
  - Circular GB: $F_{c,sat}$ = 0.61 ± 0.046, Square GB: 0.60 ± 0.036
  - Conclu etry does not have a significant effect



Effect of simulation temperature

- Current BISON model assumes $F_{c,sat}$ is independent of temperature
- Primary effect of varying temperature: gas diffusivity D_g
- Ran 5 simulations with T = 1015 K (D_g decreased by 2x)
  - Much finer microstructure at same simulation time
  - No change in calcu[illegible] = 0.60 ± 0.014

Microstructure at t = 1.98 × 10^8 s:



D_g = 0.05 nm^2/s D_g = 0.1 nm^2/s

Conclusions

- BISON predictions of fission gas release and swelling are strongly dependent on dihedral angle, surface energy, $F_{c,sat}$
  - No measured values available
- Phase-field simulations were used to calculate $F_{c,sat}$
  - Determined without needing to wait for costly post-irradiation examination
- $F_{c,sat}$ = 0.60 recommended for BISON U_3Si_2 model
- No strong effect on $F_{c,sat}$ from initial conditions, minimum bubble spacing, simulation domain geometry, temperature (in range considered)
- BISON simulations of U_3Si_2 ATR irradiation underway, using parameters determined from lower length scale calculation and their uncertainties

Thank you!

Questions?