INL/MIS-23-73088-Revision-0

Mesoscale modeling of Microstructural Evolution in High Burnup UO2 Fuel and Transient Response

June 2023

Changing the World's Energy Future

Larry Kenneth Aagesen Jr, Sudipta Biswas, Wen Jiang, Pierre-Clement A Simon, Kyle A Gamble, David Andersson, Michael Cooper, Christopher Matthews, Nathan Capps, Sophie Blondel, Brian Wirth

Idaho National Laboratory

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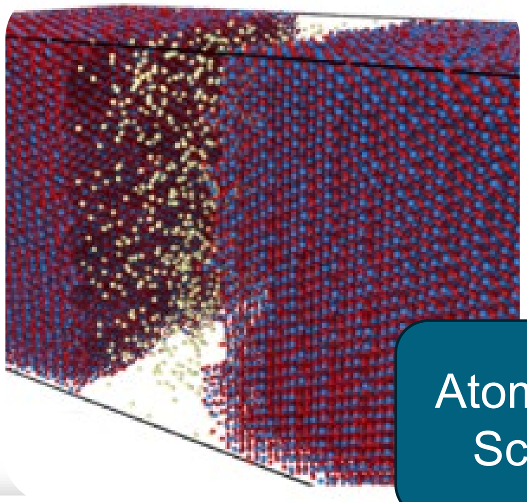
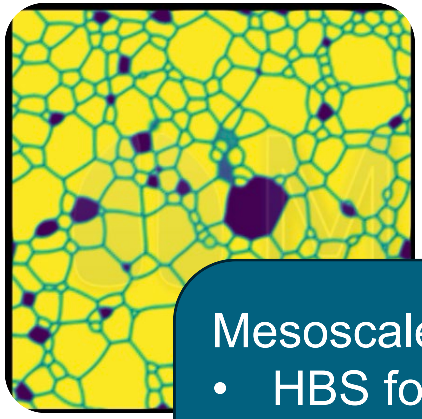
June 2023
DOE-CEA Collaboration Meeting

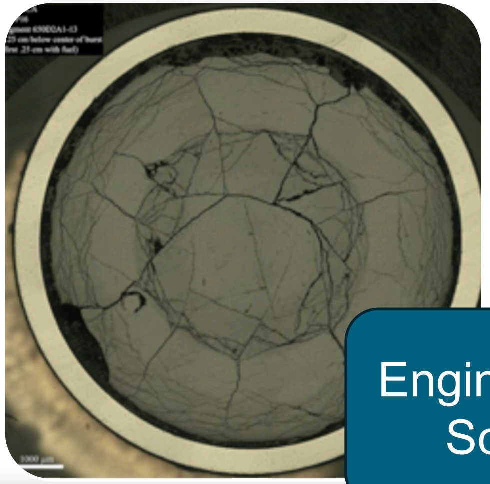
NEAMS
Nuclear Energy Advanced Modeling and Simulation

Overview

- **U.S. Industry is seeking to obtain regulatory approval to operate fuel to higher burnup**
  - Microstructure of UO_2 fuel undergoes significant changes at high burnup
  - Risk of fine fragmentation/pulverization during loss-of-coolant accident (LOCA)-type temperature transients
- **Use multi-scale simulation methods:**
  - Develop improved scientific understanding
  - Inform and parameterize engineering-scale fuel performance codes (BISON)
    - Reduce cost/time and obtain parameters that are difficult to obtain from experiment







Strategy for engineering-scale model of pulverization

- **Empirical model exists in BISON: temperature, burnup dependent**
- **Mechanistic strategy for pulverization model**
  - Track local volume fraction of HBS, gas bubble pressure in HBS region (P_g) – **evolution model**
  - Determine critical bubble pressure at which pulverization initiates (P_g^{cr}) – **fracture model**
  - Pulverization occurs when HBS volume fraction is above threshold and $P_g > P_g^{cr}$ in each element – **engineering scale**

Turnbull et al., Nuc. Sci. & Eng., 179,477 (2015).

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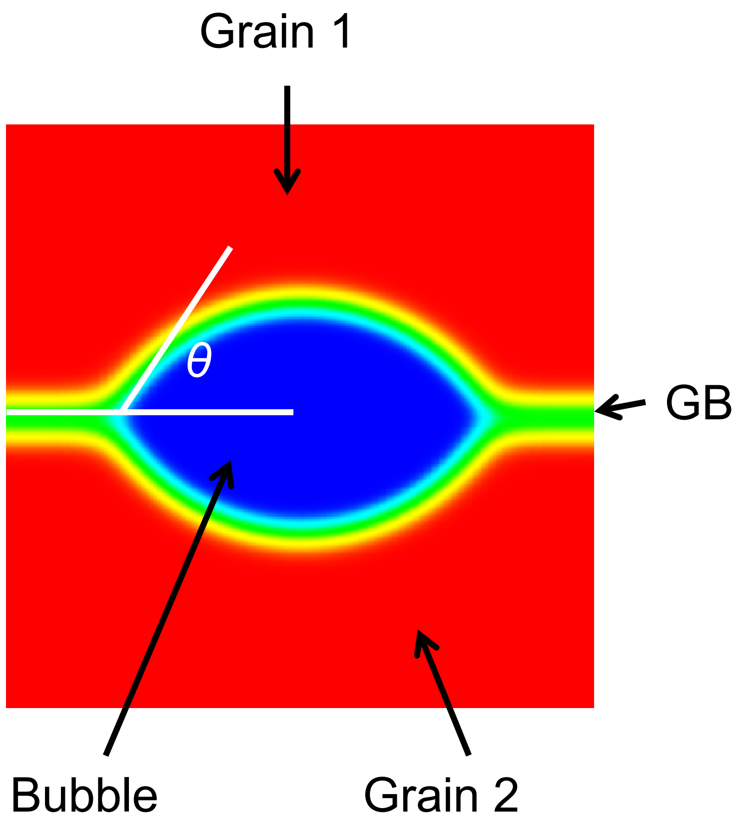
HBS formation and evolution

Phase-Field Model: Essential Physics

- **Represent intergranular bubble phase and multiple grains of UO_2**
  - Intragranular bubbles not explicitly included
- **Track vacancies and fission product species**
  - Source terms for production
- **Set interfacial energies for grain-grain and grain-bubble interfaces**
  - Controls semi-dihedral angle θ
  - Remove bulk energy contribution to interfacial energy



Phase-field Model: Microstructure

- **Represent n grains with l order parameters**
  - Use MOOSE GrainTracker so $l < n$ for large number of grains
- **Represent bubble phase with single order parameter**
- **Diffuse interface between order parameters**

Grain 0

$$\eta_{m0} = 1$$

$$\eta_{m1} = \eta_{m2} = \ldots = 0$$

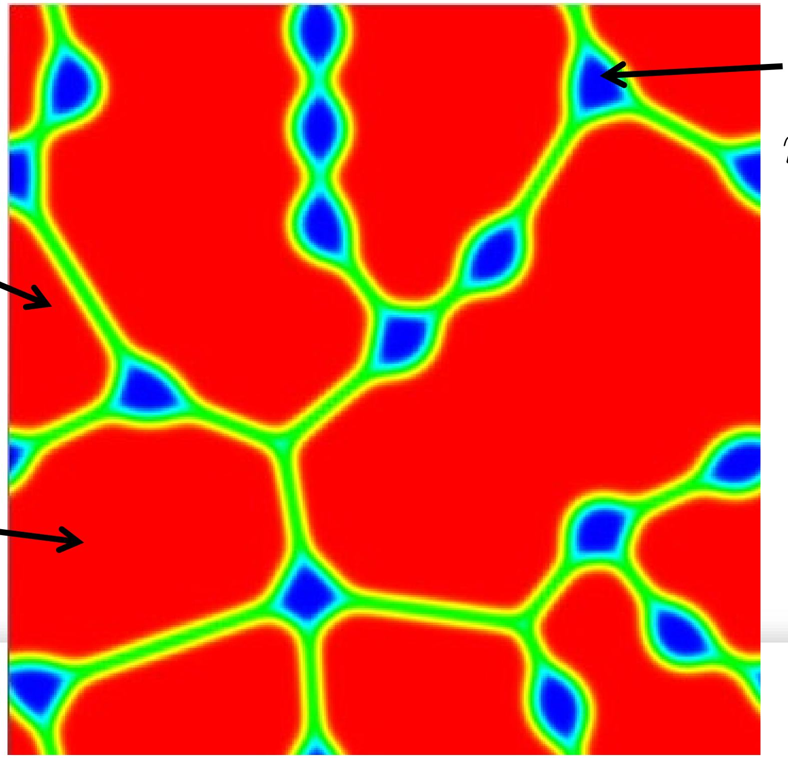
$$\eta_{b0} = 0$$

Grain 1

$$\eta_{m1} = 1$$

$$\eta_{m0} = \eta_{m2} = \ldots = 0$$

$$\eta_{b0} = 0$$



Bubble

$$\eta_{m0} = \eta_{m1} = \ldots = 0$$

$$\eta_{b0} = 1$$

Evolution equations

- **Allen-Cahn: for each order parameter,**

$$\frac{\partial \eta_{\alpha i}}{\partial t} = -L\left[m\left(\eta_{\alpha i}^3 - \eta_{\alpha i} + 2\eta_{\alpha i}\sum_{\beta}\sum_{j=1,\alpha i\neq\beta j}^{p_\beta}\gamma_{\alpha i\beta j}\eta_{\beta j}^2\right) - \kappa\nabla^2\eta_{\alpha i} + \sum_{\alpha}\frac{\partial h_\alpha}{\partial \eta_{\alpha i}}\omega_\alpha\right]$$

- **Composition: Change to chemical potential for each solute (B, C, etc.)**

$$\frac{\partial \mu_B}{\partial t} = \frac{1}{\chi_B}\left[\nabla\cdot(D_B\chi_B\nabla\mu_B) + s_B(h_\alpha, h_\beta, \ldots) - \sum_{\alpha}\sum_{i=1}^{p_\alpha}\frac{\partial \rho_B}{\partial \eta_{\alpha i}}\frac{\partial \eta_{\alpha i}}{\partial t}\right]$$

- **s_B = source term for B atoms, χ_B = susceptibility**

$$\chi_B = \frac{\partial \rho_B}{\partial \mu_B}$$

Grain Structure evolution Model

- **Initial dislocation density is a function of grain orientation**
- **Burnup dependent dislocation density evolution**
- **New grain formation observed beyond critical density**

Burnup Evolution: $\beta_{eff} = k_\beta (t - t_i)$

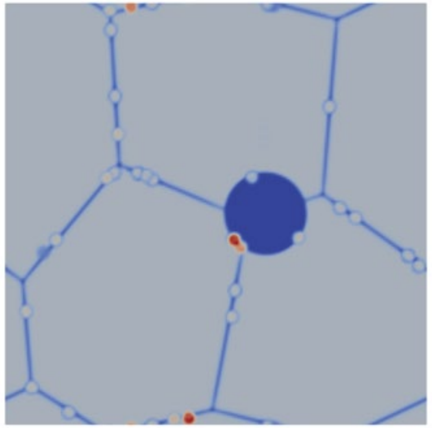
$$f_{\text{disloc}}(\eta_1, \eta_2, \ldots \eta_p) = \frac{1}{2} \mathrm{Gb}^2 \rho_{\text{eff}}(\eta_1, \eta_2, \ldots \eta_p)$$

Dislocation Density Evolution: $\log_{10} N = 2.2 \times 10^{-2} \beta_{eff} + 13.8$

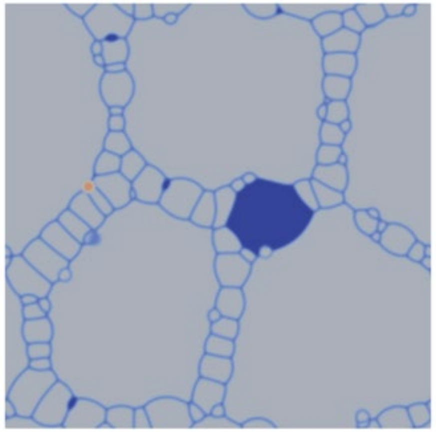
Additional Energy: $f_{disloc}(\eta_1, \eta_2, \ldots, \eta_p) = \frac{1}{2} G b^2 \rho_{eff}(\eta_1, \eta_2, \ldots, \eta_p)$

New grain formation with dislocation density buildup

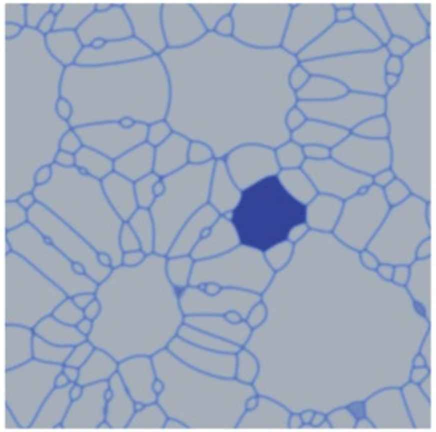
- Grain formation begins at the triple junctions, gran boundaries, and bubble surfaces
- Grain formation is followed by a coarsening stage



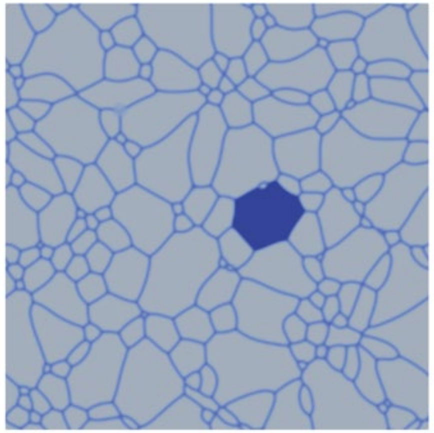
Grain Nucleation



25.17% recrystallization at 7.94×10^5 sec



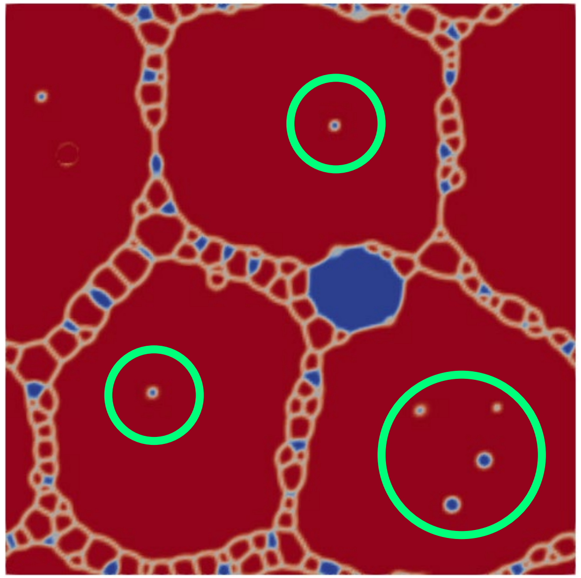
60.34% recrystallized grains at 2.24×10^6 sec



100% recrystallized grains at 2.83×10^6 sec

Limitations of phase-field-only approach

- **Doesn't resolve intragranular bubbles explicitly**
  - Not a limitation inherent to phase-field modeling, but a practical reality:
  - Need a relatively large diffuse-interface width to allow mesh to be coarse enough to simulate multiple intergranular bubbles
  - Interface width is too large to resolve intragranular bubbles at realistic size scales
  - When defect concentrations become high enough in grain interiors, intragranular bubbles do form, but at unrealistically large sizes
- **Solution: coupled phase-field/cluster dynamics approach**



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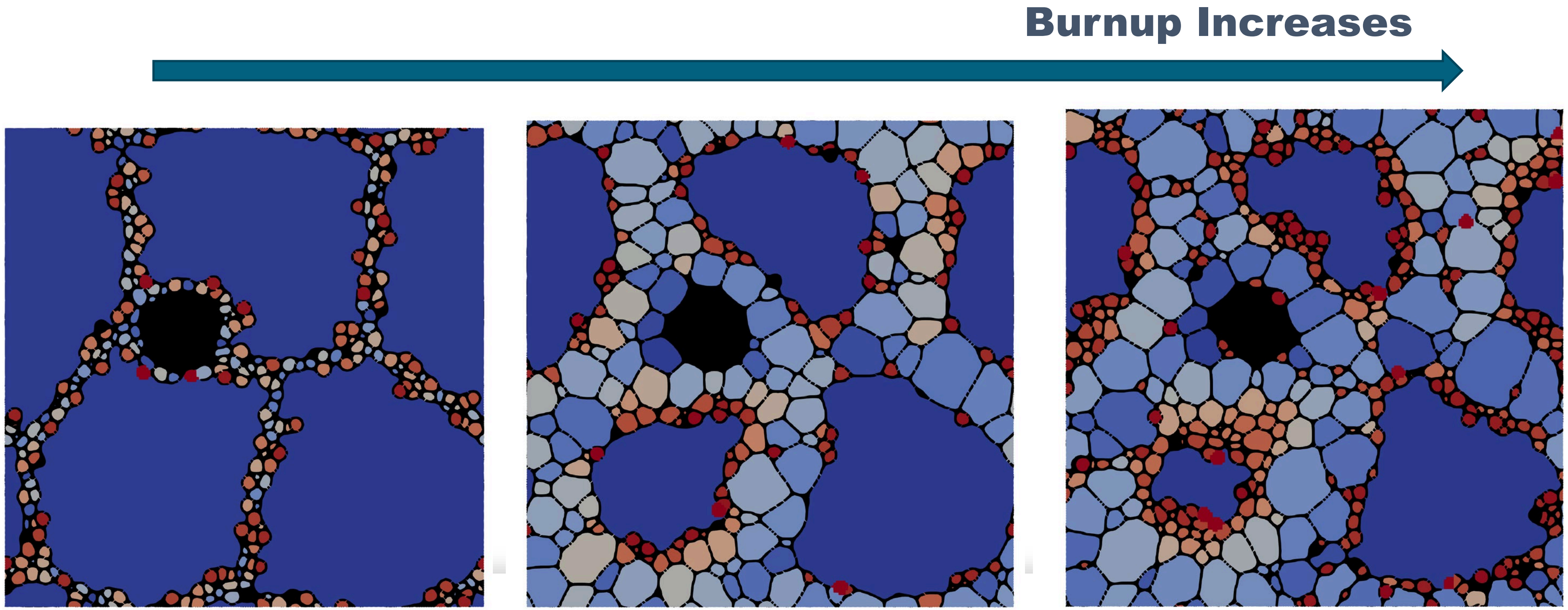
Coupled Xolotl-Marmot slides here (David)

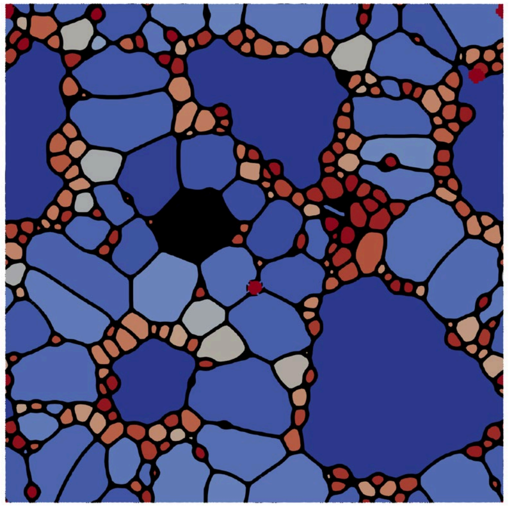
HBS Formation Using Coupled Xolotl-Marmot Model

- **Grain nucleation begins around the bubbles and triple junctions and then proceeds to grain boundaries**
- **Nucleation stage is followed by a coarsening stage**
- **Final average grain size increases with increase in temperature**

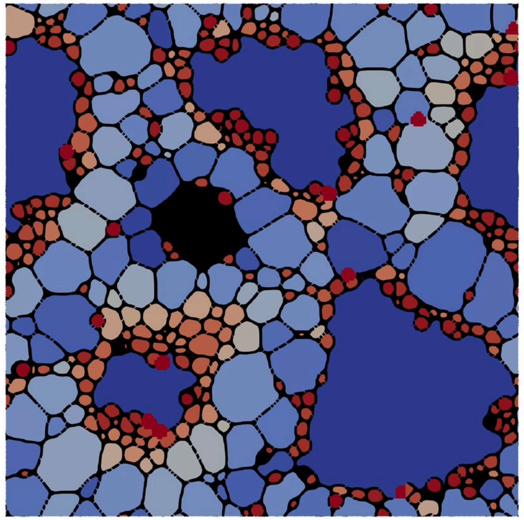


HBS Formation Using Coupled Xolotl-Marmot Model

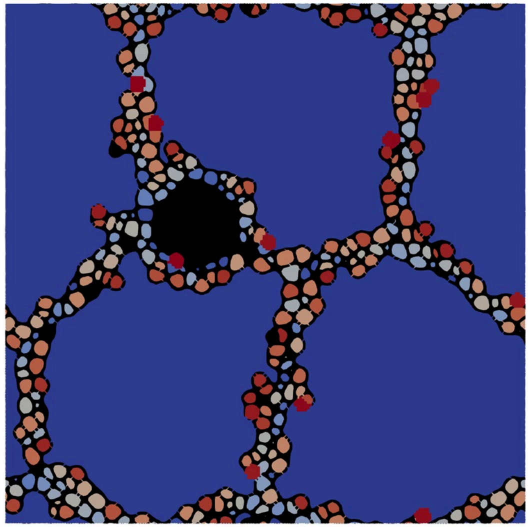
- Performed recrystallization simulations at different temperature to generate microstructures with partial HBS formation
- Grain subdivision happens in stages, new grain formation is followed by a coarsening stage



HBS formation at 1200 K



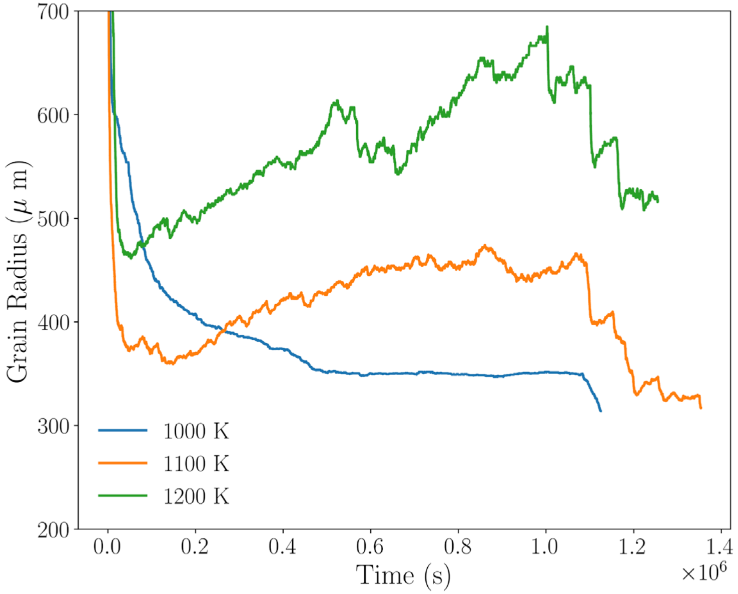
HBS formation at 1100 K

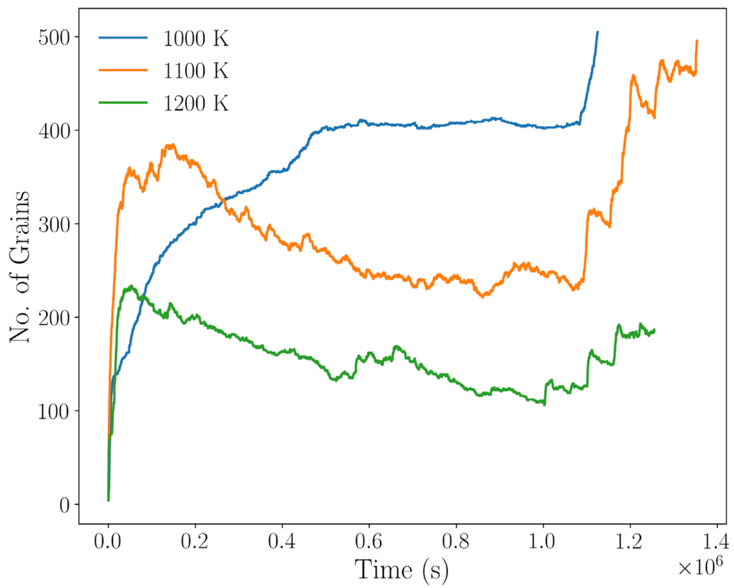


HBS formation at 1000 K

HBS Formation Using Coupled Xolotl-Marmot Model

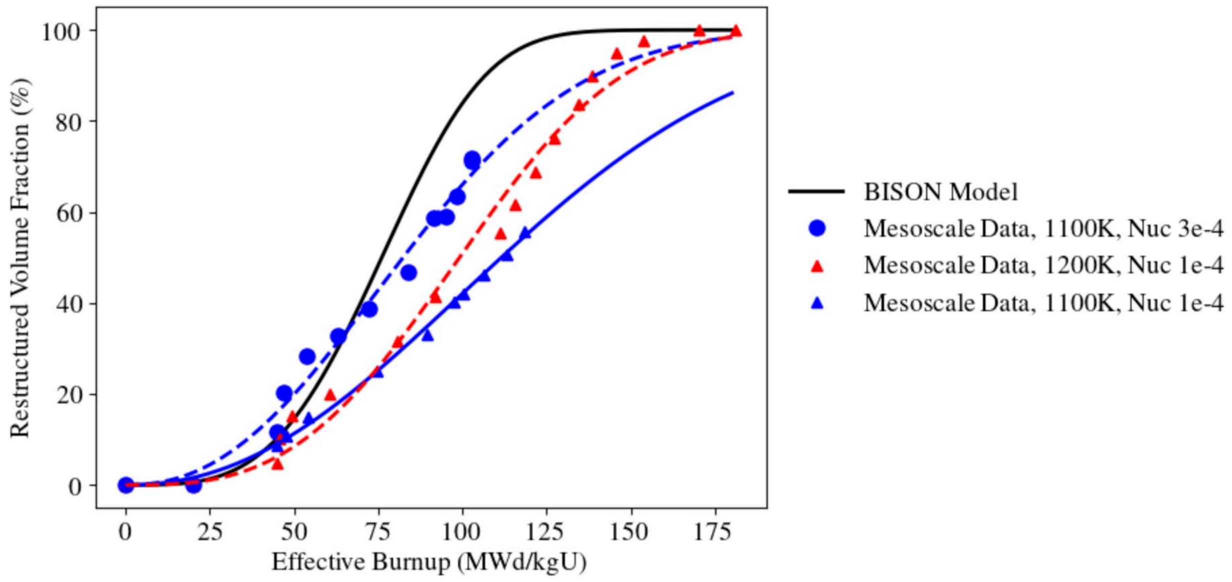
- Performed recrystallization simulations at different temperature to generate microstructures with partial HBS formation
- Grain subdivision happens in stages, new grain formation is followed by a coarsening stage





Restructuring Fraction

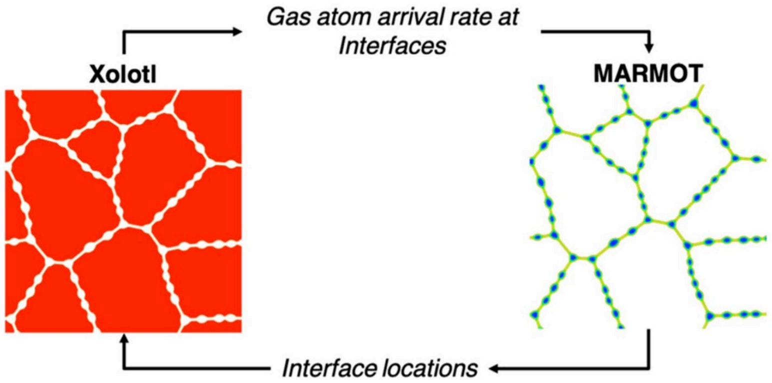
- Recrystallization fraction is calculated for different temperatures and nucleation rate
- The slope of the restructured volume fraction curve varies with temperature
- BISON restructuring fraction model will be updated based on mesoscale simulation



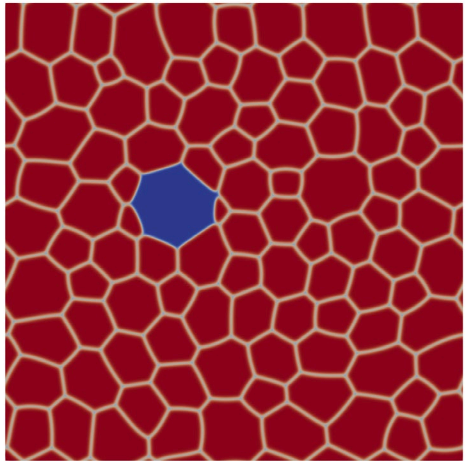
Mesoscale modeling of bubble evolution in High Burnup Structure using coupled Xolotl-Marmot Model

- Simulated bubble growth starting from a pre-existing HBS grain structure
- Fast Xe diffusivity in GBs enhances Xe transport

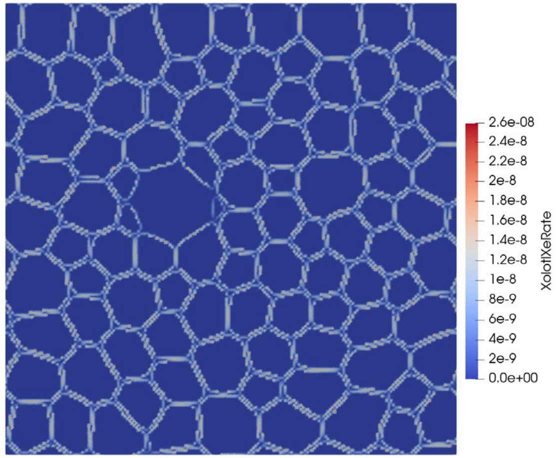
Coupled model results are consistent with hypothesized mechanism of bubble growth in HBS



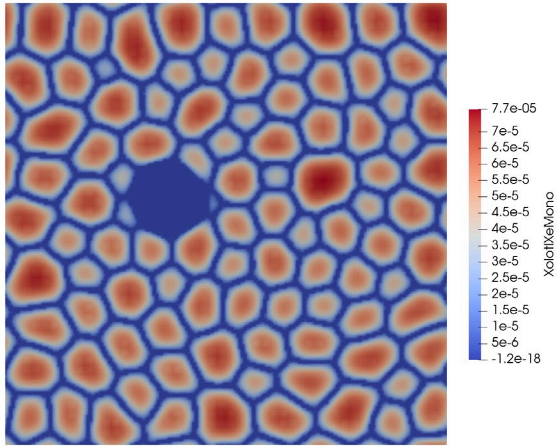
The Coupled Approach (Kim et al., Mat. Th., 2021)



Microstructural Evolution



Xe Source from Xolotl



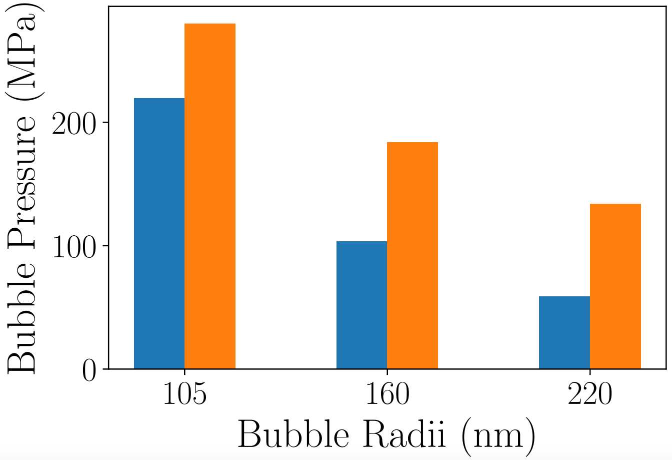
Xe Monomer Concentration

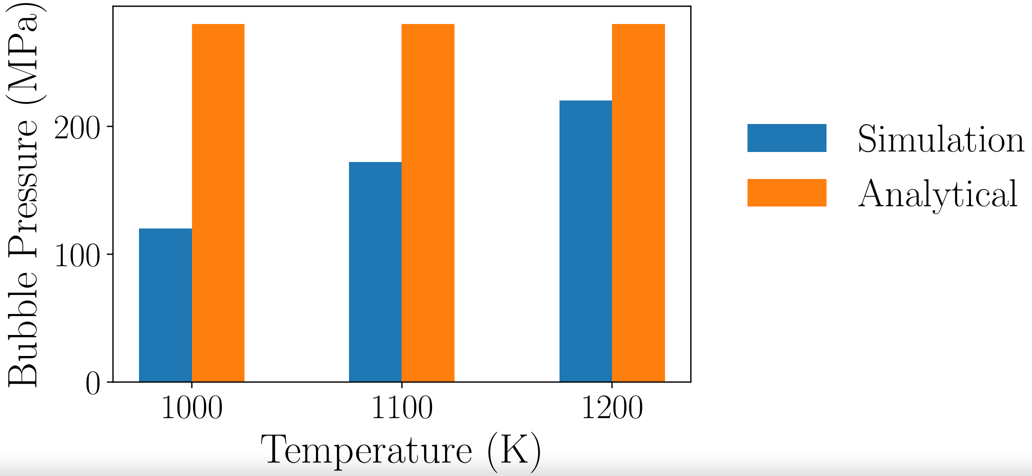
HBS Initial Bubble Pressure

- Bubble pressure is calculated from the van der Waals equation of state, $P_g = \frac{c_g k_B T}{\Omega - c_g b}$
- Pressure obtained from simulations are compared against the analytical calculation,

$$P = P_{eq} + P_{ext} = P_h + \frac{2\gamma_{st}}{r} + \frac{Gb}{r}$$

Model underpredicts the pressure, safe to use analytical dislocation punching pressure for initial bubble pressure in BISON pulverization criterion



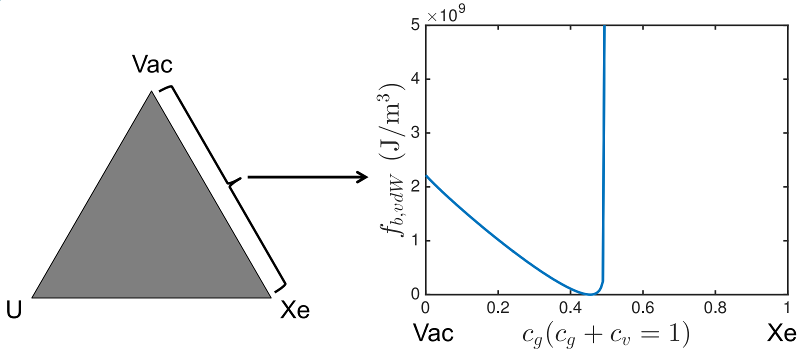


HBS formation: parameterization for lower temperatures

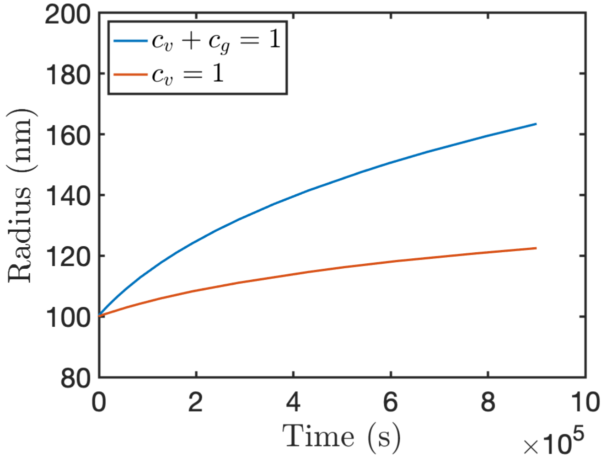
- Atomistic calculations have shown Xe interstitials are most mobile Xe species at lower temperatures (<1200 K)
  - Previous phase-field modeling was parameterized assuming substitutional Xe, enforced $c_v + c_g = 1$
- Account for interstitial Xe: instead enforce $c_v = 1$
- Update free energy of gas phase:

$$f^b_{chem} = c^b_g \frac{kT}{V_a}\left[\ln\left(\frac{1}{n_Q(\frac{V_a}{c^b_g} - b)}\right) - 1\right] + \frac{k_p}{2}(1 - c^b_v)^2 + f_0$$

- Tested phase-field implementation to ensure correct compositions in comparison with numerical calculation of equilibrium
- Tested growth in KKS+surface tension model*
  - Growth rate decreased for interstitial Xe



Gas phase parameterization for UO_2 assuming substitutional Xe



Bubble growth rate

*Aagesen et al., JNM, 557, 153267 (2021)

HBS formation: Xe Equation of State

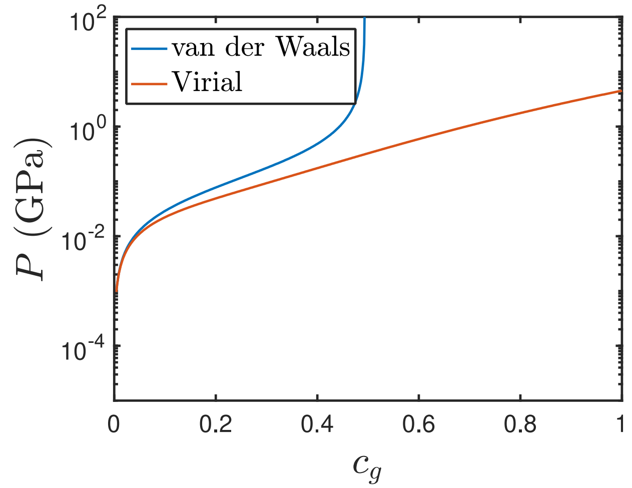
- Past phase-field and BISON modeling has often used van der Waals equation of state (EOS) for Xe
  - Pressure-volume relationship, Helmholtz free energy
  - Breaks down as assumed hard-sphere Xe volume is approached
- Using new EOS developed based on MD*, in virial form:

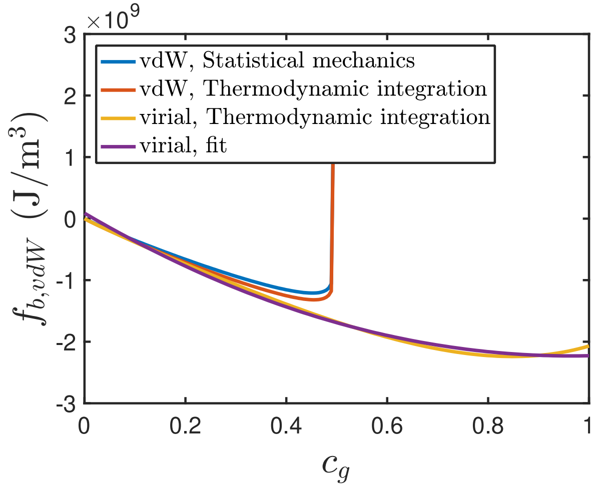
$$P = \rho kT\left(1 + B\rho_r + C\rho_r^2 + E\rho_r^4\right),$$

- Performed thermodynamic integration to obtain Helmholtz free energy from $P - \rho$ relationship:

$$f_{b,vdW} = -\int_{v_g^0}^{v_g} P(v_g)dv_g \qquad v_g = N_a/\rho$$

- Fit parabola to Helmholtz free energy for convenience to include in phase-field modeling





T = 700 K

*Yang & Wirth, JNM, 572, 154089 (2022)

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Transient response in HBS (rim)

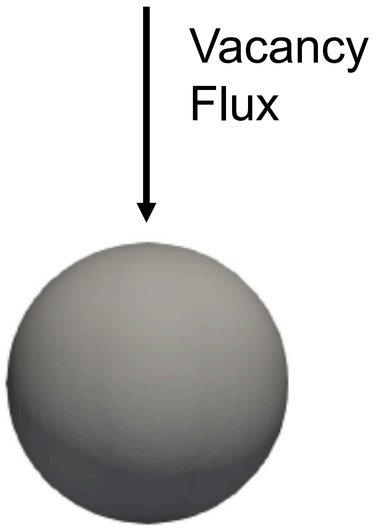
Strategy for engineering-scale model of pulverization

- **Empirical model exists in BISON: temperature, burnup dependent**
- **Mechanistic strategy for pulverization model**
  - Track local volume fraction of HBS, gas bubble pressure in HBS region (P_g) – **evolution model**
  - Determine critical bubble pressure at which pulverization initiates (P_g^{cr}) – **fracture model**
  - Pulverization occurs when HBS volume fraction is above threshold and $P_g > P_g^{cr}$ in each element – **engineering scale**

Turnbull et al., Nuc. Sci. & Eng., 179,477 (2015).

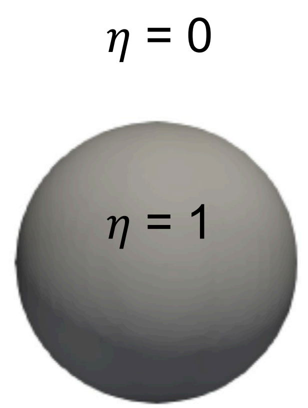
HBS bubble response to LOCA transient

- Bubbles in HBS region are ~1 µm and believed to be overpressurized relative to equilibrium (based on observed dislocation punching around bubbles):
  - $P = \frac{2\gamma_{st}}{R} + \sigma_H$
- Overpressurized bubbles exert compressive stress in the radial direction on the surrounding matrix.
- During LOCA transient, temperature and therefore bubble pressure increases further, causing stress in the matrix to increase further. Compressive stress leads to increased vacancy flux to bubble, causing bubble growth.
- Key Questions:
  - Does significant bubble growth occur during duration of a LOCA transient? If so, need to account for size change in fracture model.
  - What is the pressure response to a given temperature transient?



Phase-field model: Essential physics

- **Single order parameter η to represent gas bubble and fuel matrix phase**
  - Current model does not consider grain boundaries
- **Track vacancies and fission product gas atoms**
  - Use Xe properties for fission product gases
  - Source terms for production, sink term to limit vacancy concentration to steady-state
- **Chemical and elastic energy contributions**
- **Solid-bubble interfacial energy**
  - Kim-Kim-Suzuki (KKS) approach to remove bulk energy contribution to interfacial energy
- **Surface tension of bubble-matrix interface**
- **Xe gas pressure**



Evolution equations

- **Allen-Cahn for order parameter:**

$$\frac{\partial \eta}{\partial t} = -L\left(\frac{\delta F}{\delta \eta}\right)$$

$$= L\left[\frac{dh}{d\eta}\left[(f_T^m - f_T^b) - \mu_v(c_v^m - c_v^b) - \mu_g(c_g^m - c_g^b)\right] - W\frac{dg}{d\eta} + \kappa\nabla^2\eta\right]$$

- **Cahn-Hilliard for vacancy and gas concentration (source for vacancies and gas atoms, sink for vacancies to approximate recombination):**

$$\frac{\partial c_v}{\partial t} = \nabla \cdot M_v \nabla \mu_v + s_v - K_v c_v^m$$

$$\frac{\partial c_g}{\partial t} = \nabla \cdot M_g \nabla \mu_g + s_g$$

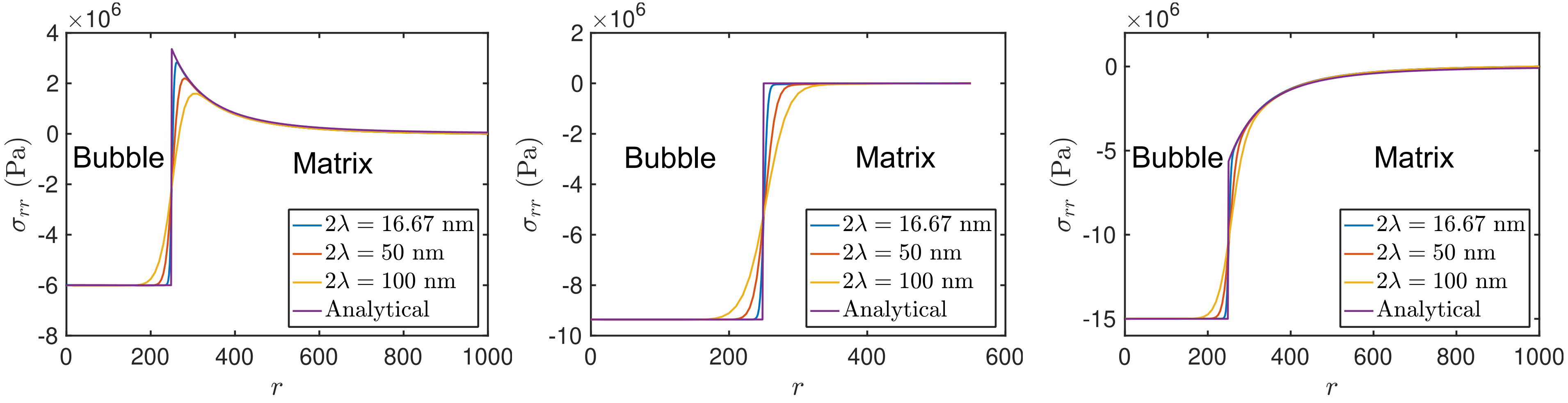
- **Mobilities are a function of defect diffusivities:**

$$M_g = \frac{hD_g^b + (1-h)D_g^m}{d^2 f/dc_g^2} \qquad M_v = \frac{hD_v^b + (1-h)D_v^m}{d^2 f/dc_v^2}$$

- **+ KKS system constraints, mechanical equilibrium equation including surface tension**

Phase-field model verification and testing

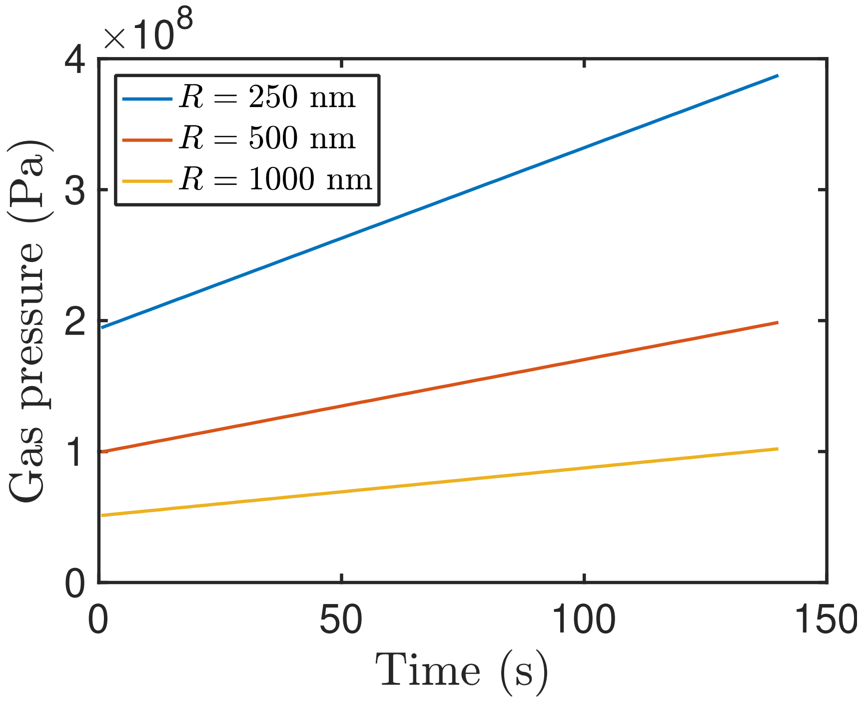
- **Considered stress state in equilibrium, underpressurized, overpressurized bubbles (1D simulation in radial coordinates)**
  - Equilibrium: σ_{rr} = 0 in surrounding solid matrix
  - Under/overpressurized: σ_{rr} = +/-, corresponding to tensile/compressive stress in surrounding matrix
  - Converge to analytical solution as interface width (2λ) decreases



Underpressurized | Equilibrium | Overpressurized

Bubble response during LOCA transient

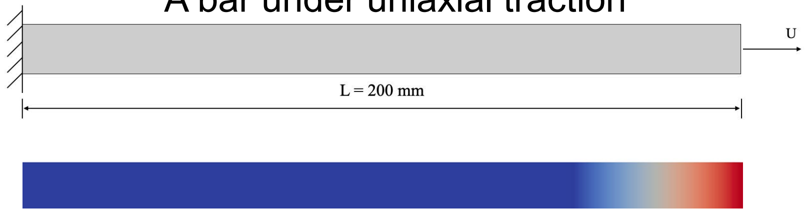
- **Vary initial radius: 250 nm, 500 nm, 1000 nm**
  - Change domain size to maintain 10% porosity
- **Initial pressures set at upper bound estimate from dislocation punching: 200, 100, 50 MPa.**
- **No significant change in bubble size**
  - Similar calculations for varying initial bubble pressure, external pressure also showed no significant change in bubble size
- **Based on these results, do not need to consider bubble size change in fracture model**

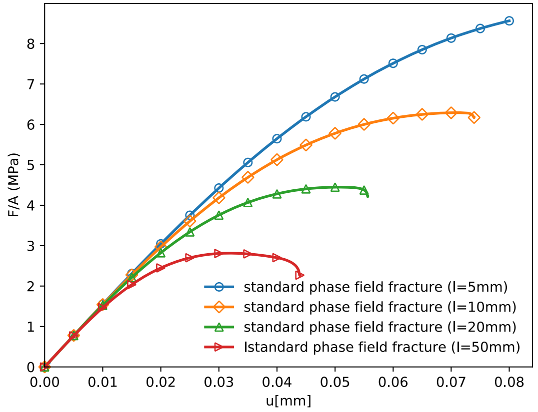


Phase-field fracture simulation of HBS fuel

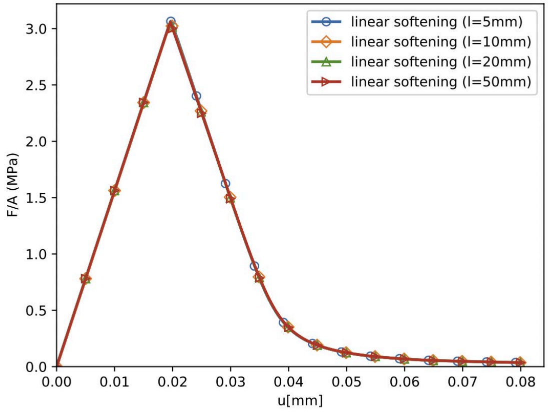
- Developed phase-field model for quasi-brittle fracture
  - It remains elastic behavior before crack initiates
  - Critical fracture strength is independent of length-scale parameter
  - It can predict general softening laws, such as linear softening law
- Developed phase-field model for pressurized fracture
  - The pressure is applied on regularized fracture surfaces.
  - The presence of pressure on fractures will change the behavior of crack propagation: from stable to unstable.

A bar under uniaxial traction

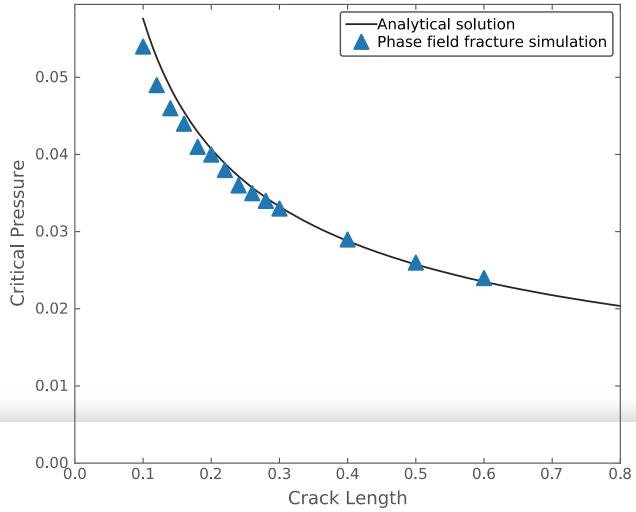




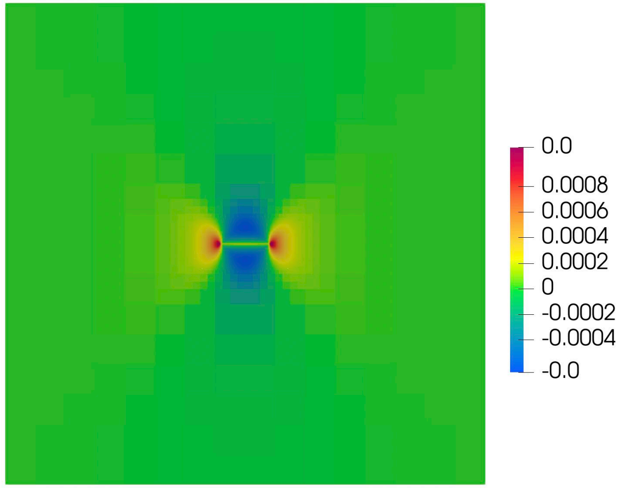
Classical brittle model



Quasi-brittle model



critical pressure vs. crack length



Stress contour of a pressurized crack

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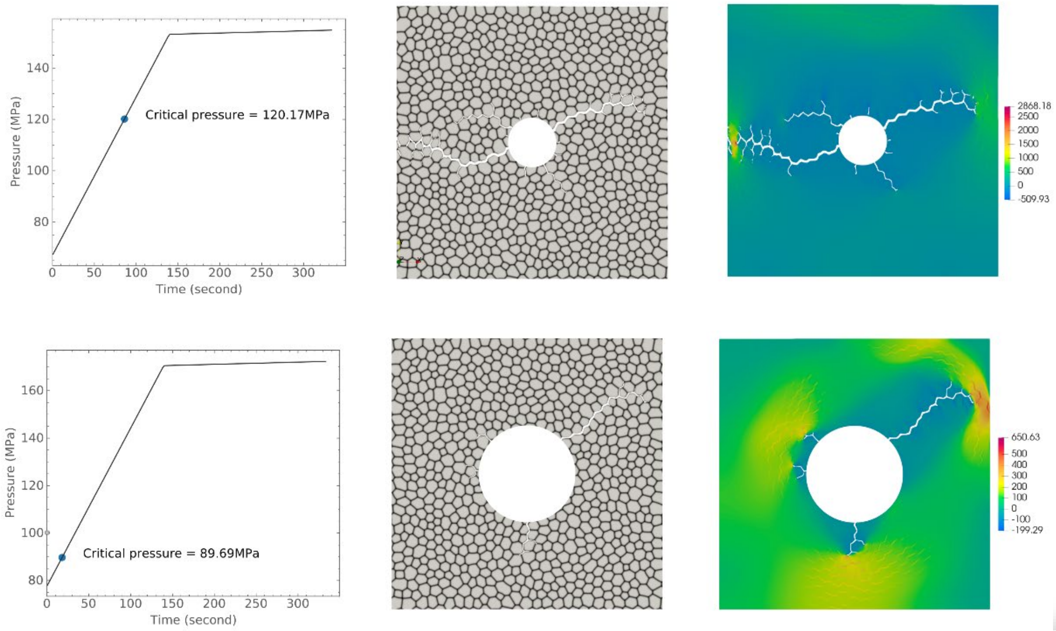
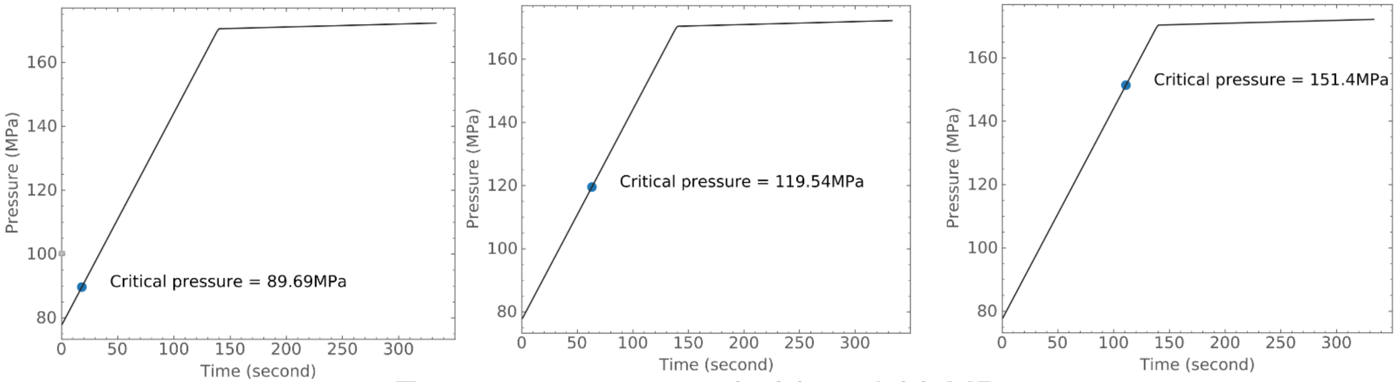
Phase-field fragmentation modeling

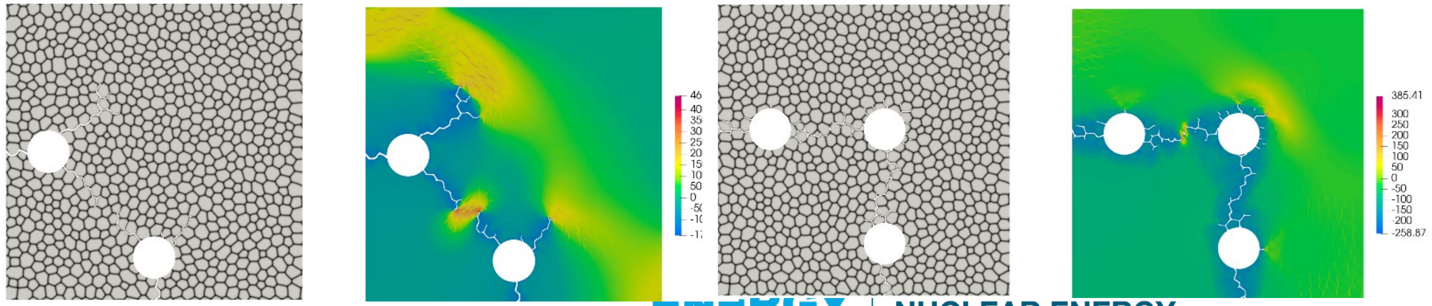
- **Phase-field fracture model was used to study HBS fragmentation behaviors**
  - Effect of bubble sizes: critical pressure is lower for the larger bubble
  - Effect of external pressure: critical pressure becomes higher for larger external pressure values
  - Effect of bubble interaction: fragmentation size is likely determined by bubble spatial distribution



Bubble size = 0.25 μm and 0.5 μm



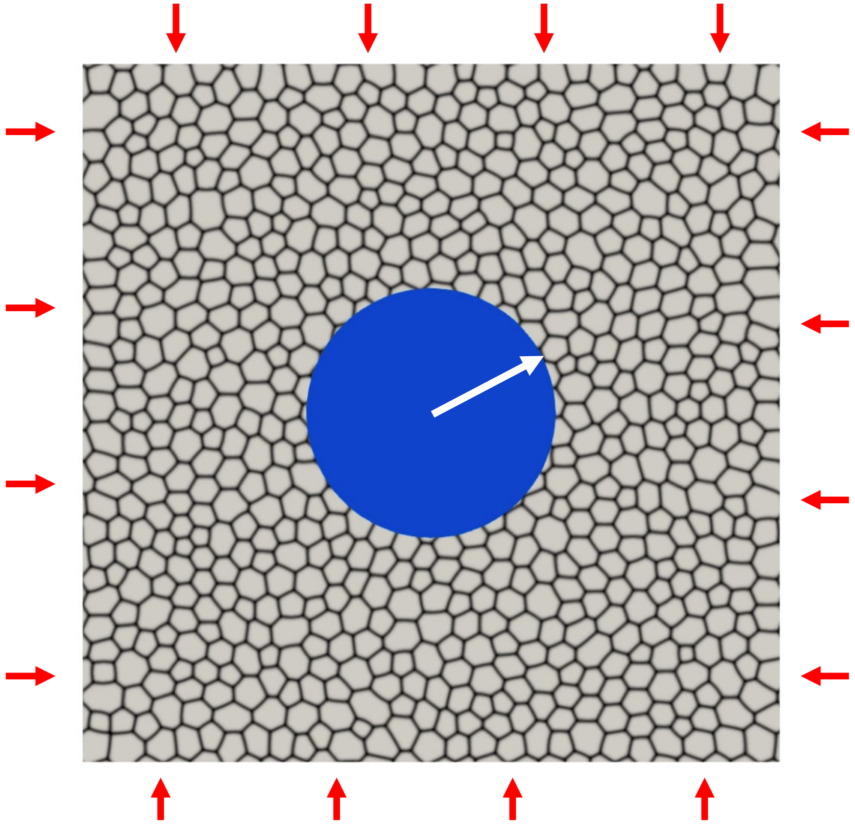
External pressure = 0, 30 and 60 MPa

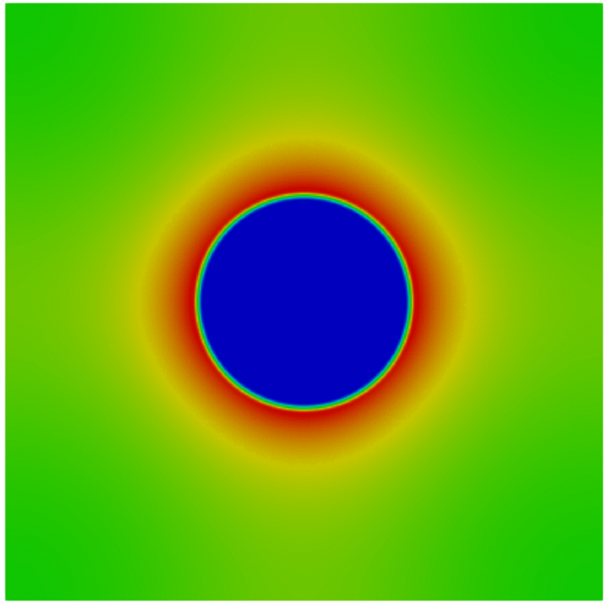


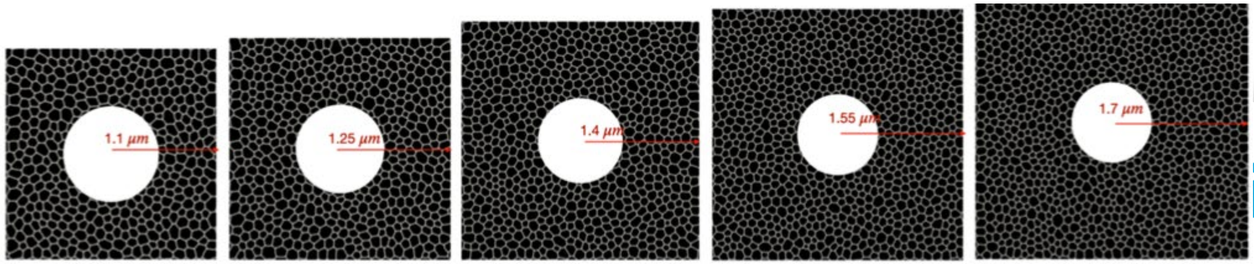
Two and three bubbles interaction

Determine BISON pulverization criterion

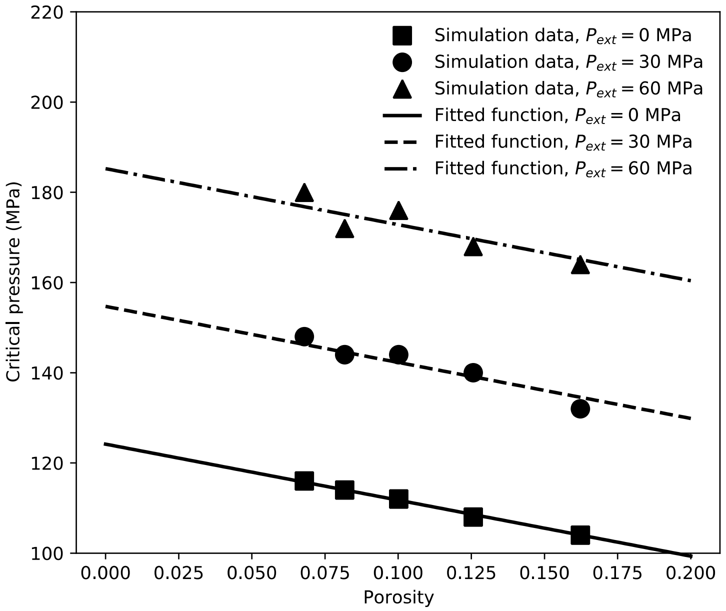
- **Phase-field fracture model was used to inform pulverization criterion for BISON models**
  - Use periodic boundary conditions to account for multi-bubble interaction.
  - Consider varying porosity (p), external pressure (p_{ext}), critical fracture stress of grain boundary (σ_c)

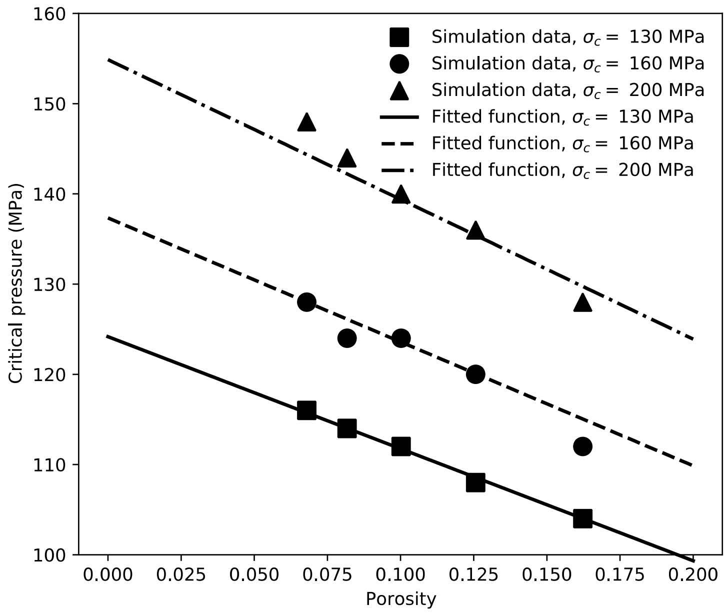






BISON pulverization criterion





- **Function to fit the data (a, b, c are fitting constants) :**
  - $P_g^{cr} = [a + b(\sigma_{cr} - 130)](1 - p) - cP_{ext}$
- **Implemented in BISON. Validation with BISON assessment cases is in progress**

BISON validation: Studsvik Rods 191 and 196

- **The Studsvik tests were a series of experiments conducted by NRC at the Studsvik reactor.**
- **Rods 191 and 196 were initially added to Bison's validation suite as part of an EPRI project.**
- **Investigate the impact on predicted fuel release during the LOCA transient for the existing empirical Turnbull pulverization threshold and the analytical and phase-field informed lower length scale thresholds.**
- **Initial predictions from phase-field based pulverization criterion are same order of magnitude as empirical model**
  - Mesh convergence studies are in progress

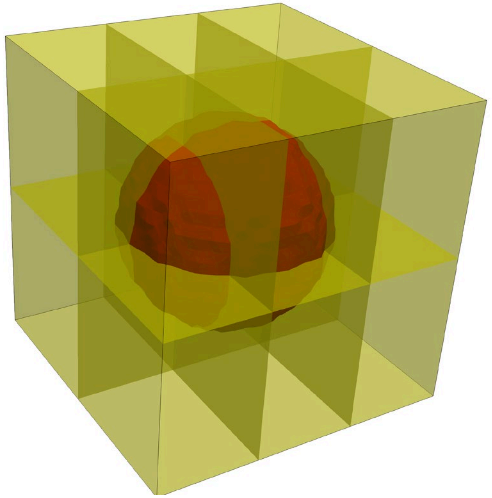
Rod 191		Rod 196	
Model	Mass Released (g)	Model	Mass Released (g)
Empirical	18.6	Empirical	6.56
Analytical	0	Analytical	0
Phase-field	25.4	Phase-field	16.2
Experiment	> 41	Experiment	0

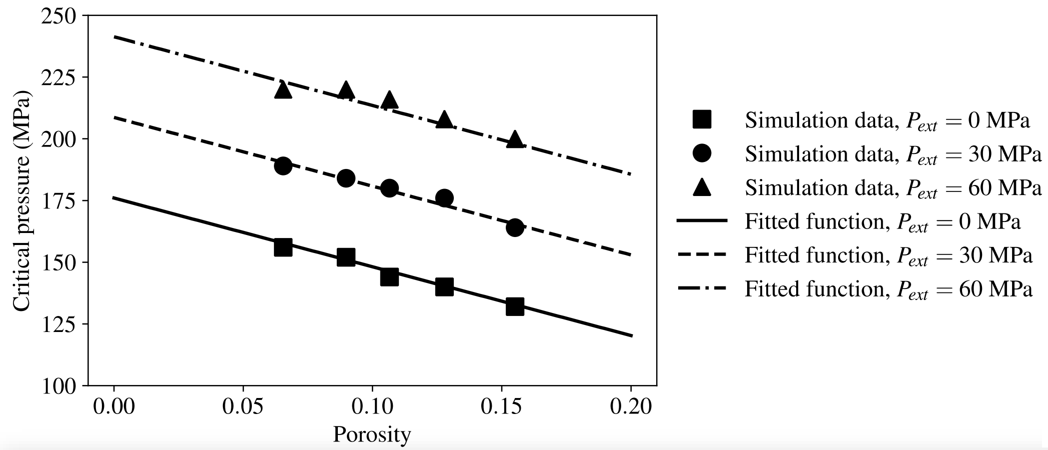
Test ID	189	191	192	193	196	198
Rod ID	AM2-E08-2-1	AM2-F10-2-2	AM2-E08-2-2	AM2-F10-2-1	M14-L3	M14-L2
Comments	Ramp to rupture test	Ramp to PCT, held for 25 s at PCT	Ramp to PCT, held for 5 s at PCT	Ramp to PCT, held for 85 s at PCT	Ramp to rupture test	Ramp to PCT, held for 85 s at PCT
Cladding	ZIRLO	ZIRLO	ZIRLO	ZIRLO	ZIRLO	ZIRLO
Rod Type	UO_2	UO_2	UO_2	UO_2	IFBA - ZrB_2 coating	IFBA - ZrB_2 coating
Burnup (GWd/MTU)	≈ 72	≈ 71	≈ 72	≈ 71	≈ 55	≈ 55
Adjacent Hydrogen Measurement (wppm)	176	271	288	187	149	<149
Cladding OD (mm)	9.5	9.5	9.5	9.5	9.14	9.14
Cladding thickness (mm)	0.57	0.57	0.57	0.57	0.57	0.57
PCT (°C)	950 ± 20	1160 ± 20	1160 ± 20	1160 ± 20	960 ± 20	1160 ± 20
Max. Burst Strain (%)	48	50	56	51	25	25
Fill Pressure (bar)	110	110	82	82	82	82
Rupture Pressure (bar)	113	104	77	77	72	74
Rupture Temperature (°C)	700	680	700	728	686	693
Rupture Opening Width (mm)	10.5	17.5	9.0	13.8	0.2	1.6
Rupture Opening Axial Length (mm)	23.9	21.6	22.7	17.8	1.5	11.0
Fuel Mass Released During LOCA (g)	>41	52	68	105	0	0
Fuel Mass Release TOTAL (g)	>61	59	84	110	77	62
Measured "Empty" Length (mm)	148	125	165	205	157	131

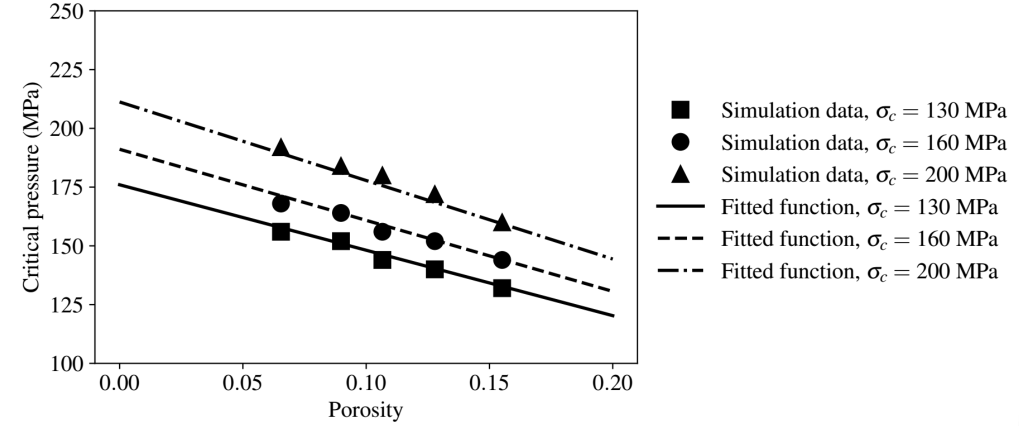
Phase-field Fracture Simulations

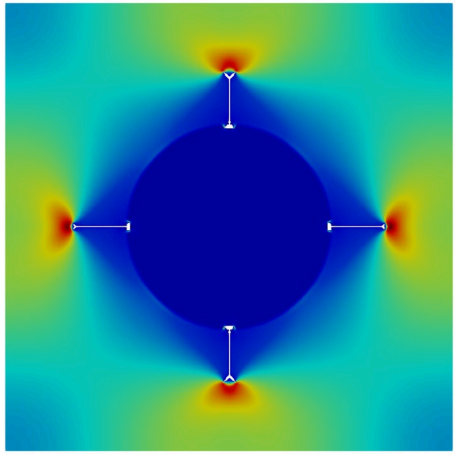
- Using simplified 3D geometry to generate the pulverization criteria
- BISON pulverization criteria is updated based on 3D phase-field fracture simulations

3D Critical pressure is higher than 2D simulations



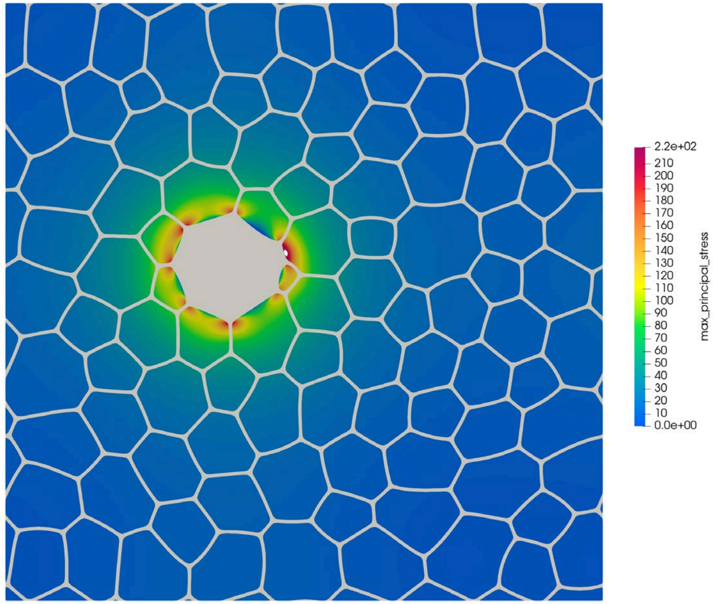


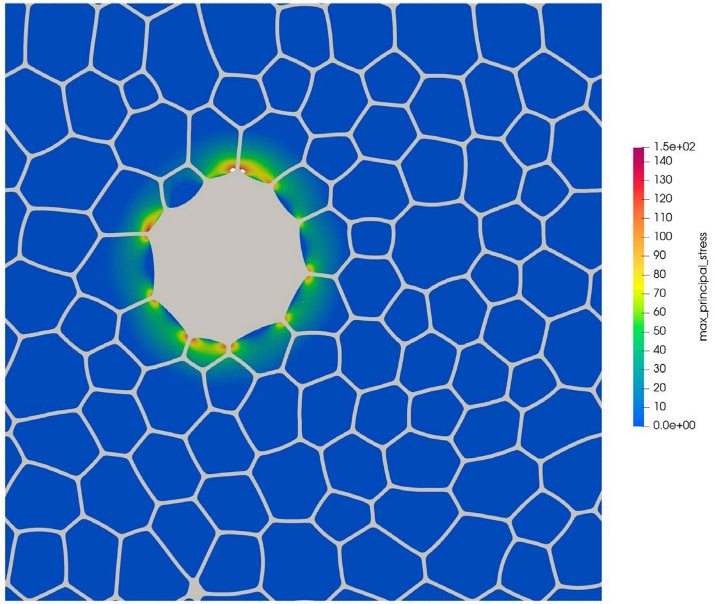




Effect of Bubble Geometry

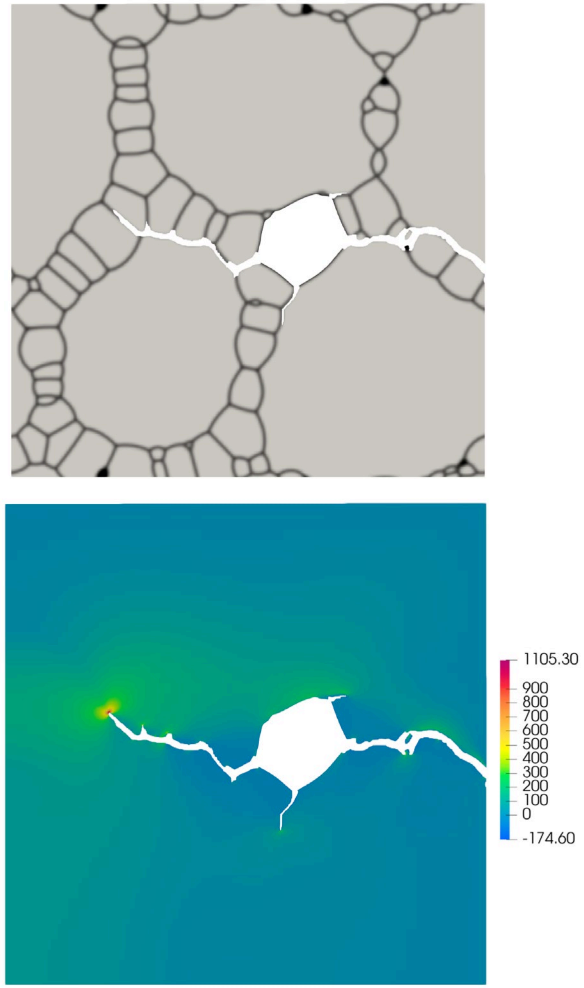
- Pulverization criteria is defined by the critical stress for crack initiation
- Bubble geometry plays important role in crack initiation
- Stress concentration is observed around bubble notches



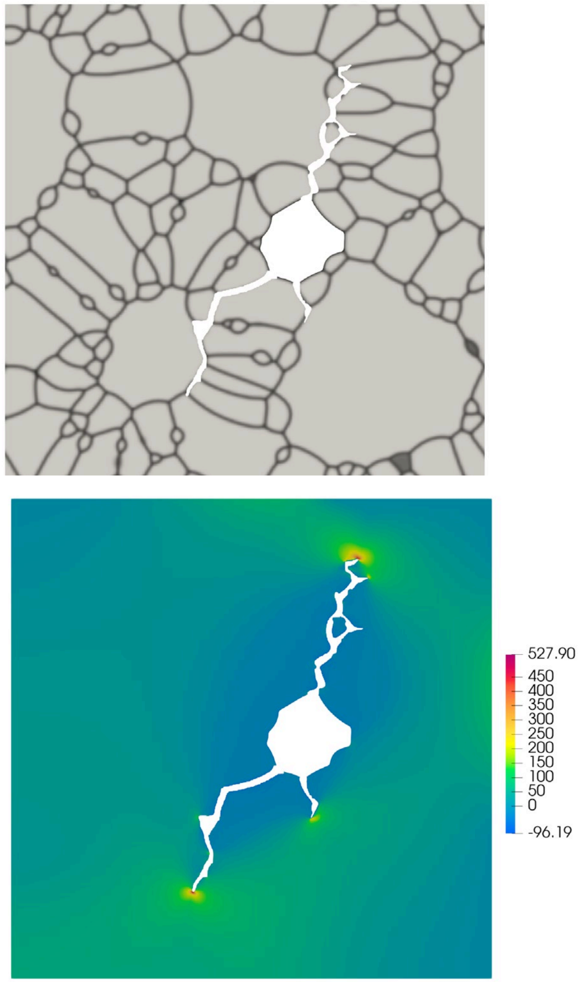


Fragmentation in partial HBS

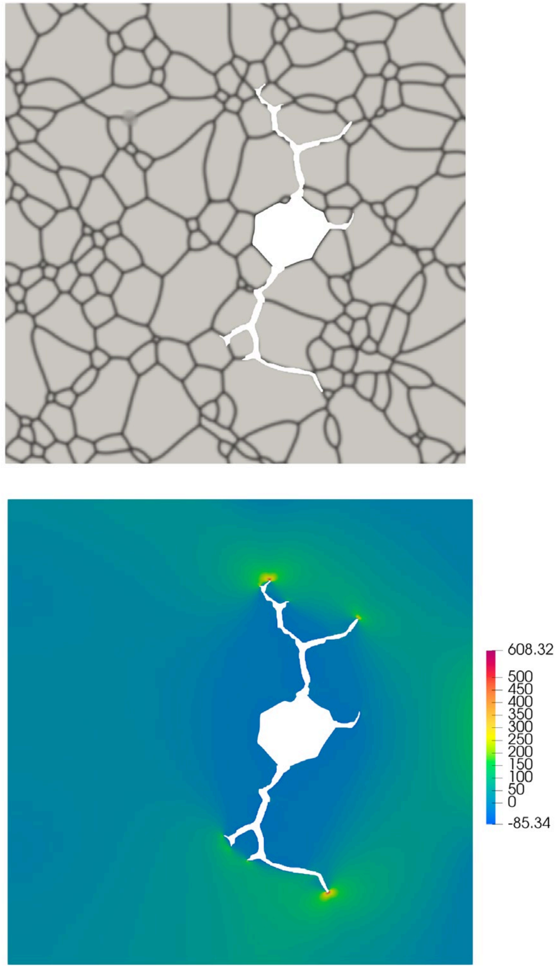
- **Phase-field fracture model was used to simulate fragmentation behaviors of partial HBS**
  - Use the output from the HBS formation simulations as our initial condition
  - Three HBS at different recrystallization stages with 25%, 60% and 100% recrystallization fraction were considered
  - Crack initiation locations and crack propagation paths varied among the three cases because recrystallized grain structures change.



25 % recrystallization stage



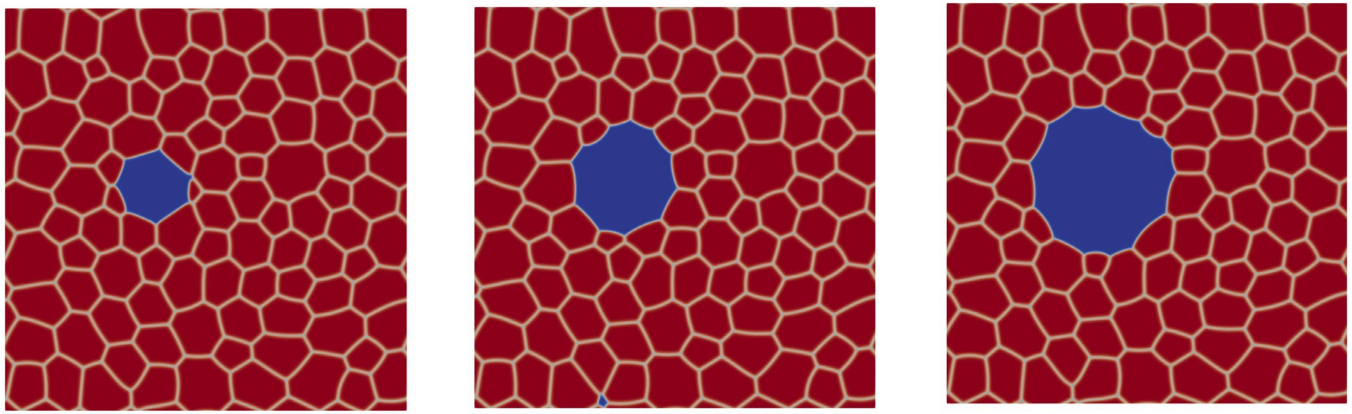
60 % recrystallization stage



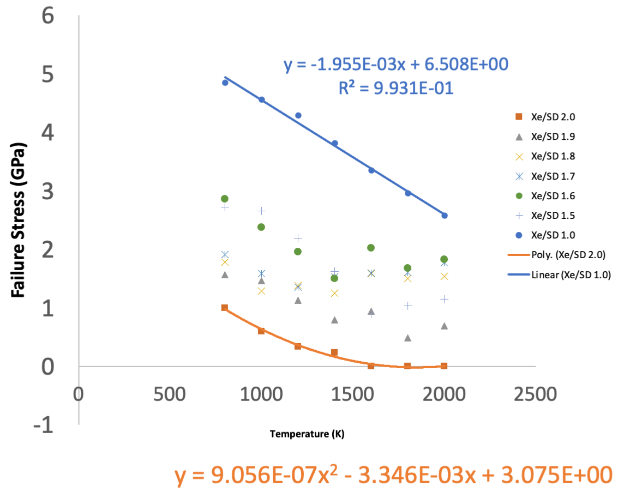
100 % recrystallization stage

Phase-field Fracture Simulations: Model Update

- HBS microstructures generated through phase-field simulations are used as initial conditions
- Updated the failure stress values based on MD data
- Temperature is ramped from 700 K at 10 K/s ramping rate, thermal strain is included



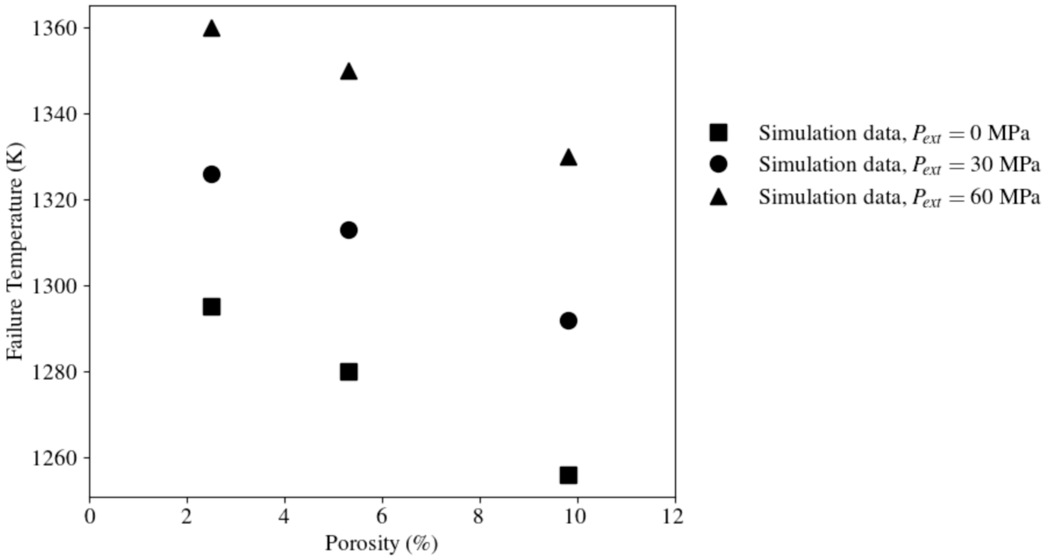
Initial conditions with different porosities

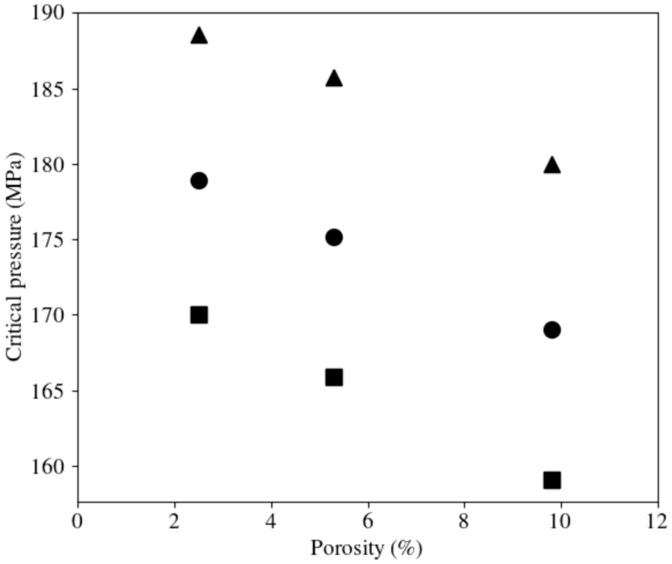


Failure Stress from MD Simulations

Critical Stress for Pulverization

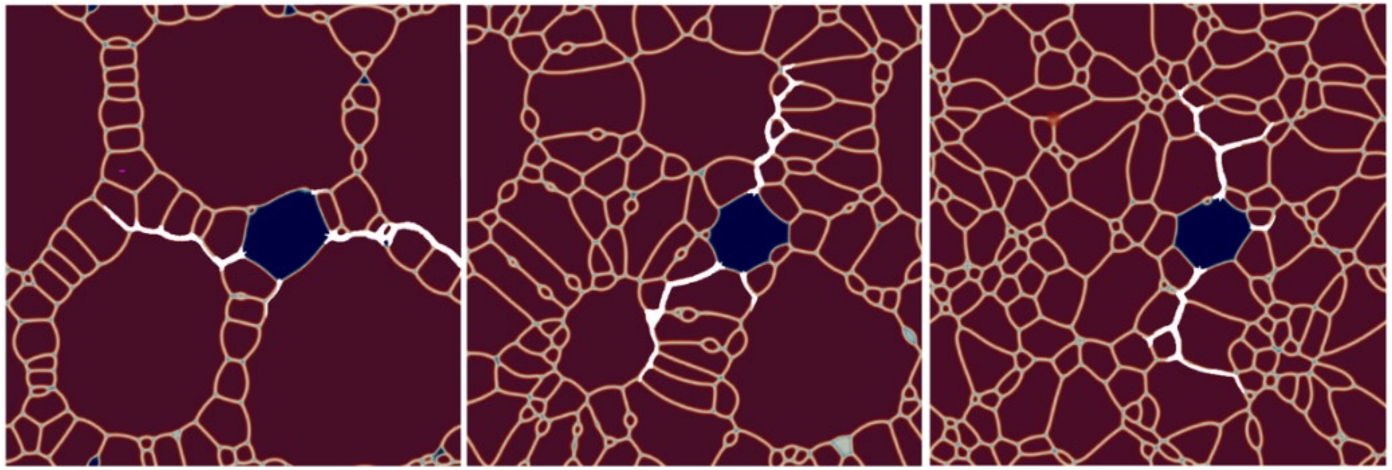
- Pulverization is caused by a combination of factors, reduced critical stress, increased temperature and bubble pressure
- Failure is observed around 1250K-1360K temperature range, external
- HBS microstructures generated through phase-field simulations are used as initial conditions





Future Work

- Resolve issues with the low temperature mesoscale simulations
- Validate the model predictions based on experimental observations
- Propagate uncertainties between length scales
- Update BISON criteria for restructuring fraction and pulverization
- Complete BISON validation with available experimental data



Fracture Progress in HBS

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Questions?

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