INL/MIS-22-66481-Revision-0

Summary of physics-based pulverization model development for high-burnup fuel

March 2022

Changing the World's Energy Future

Larry K Aagesen Jr, Sudipta Biswas, Wen Jiang, Kyle A Gamble, Michael Cooper, Topher Matthews, David Andersson

Idaho National Laboratory

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Summary of physics-based pulverization model development for high-burnup fuel

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U.S. DEPARTMENT OF ENERGY

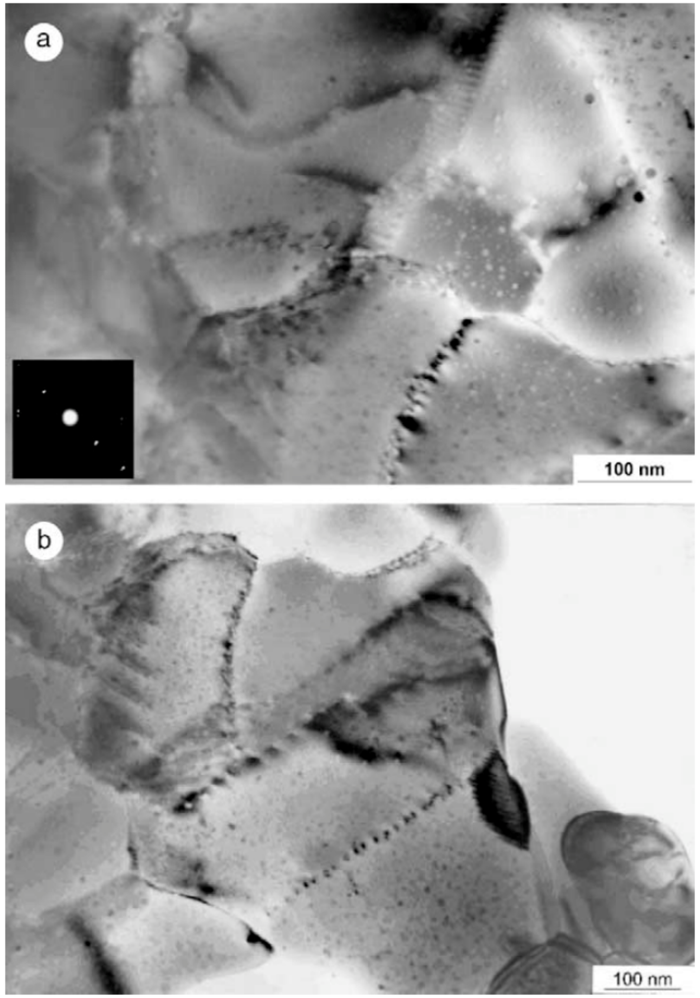
Overview

- Preliminary model implemented in BISON to predict pulverization in response to Loss of Coolant Accident (LOCA)-type temperature transient
- Atomistic work
  - Nanometer-size bubble pressure evolution
  - Impact of nanometer-size bubbles on grain boundary strength
- Mesoscale work
  - Calculation of pressure in High Burnup Structure (HBS) bubbles
  - Response of HBS bubbles to LOCA transient
  - Simulations of fracture in HBS region driven by LOCA transients
- Multi-scale approach tying this work together to an engineering-scale model
- Future plans to improve the model

Atomic scale simulation of fragmentation mechanisms

- HBS formation is characterized by coarsening of bubbles to micron-sized and sub-grain formation
- Experimental observation of nm-size bubbles decorating sub-grains (Sonoda et al., see right)
- Over-pressurized bubbles have been suggested as cause of fragmentation/pulverization of HBS during temperature ramps
- Hypothesis - the nm-size bubbles can become over-pressurized and then drive pulverization:
    - During steady-state, can the bubbles become highly pressurized due to irradiation processes of Xe interstitial diffusion? Assess this using cluster dynamics and phase field simulations as part of NEAMS
    - During a temperature ramp, can high-pressure small bubbles at grain boundaries cause de-cohesion? Initially EPRI-funded now NEAMS

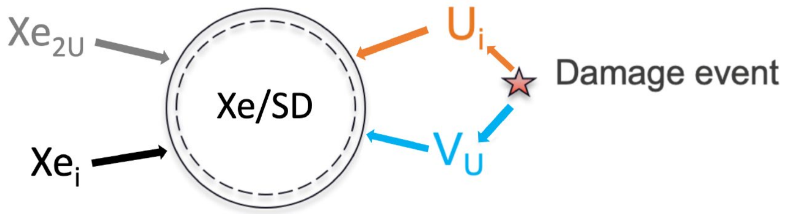
Experimental observation of nm-size bubbles at sub-grains in HBS

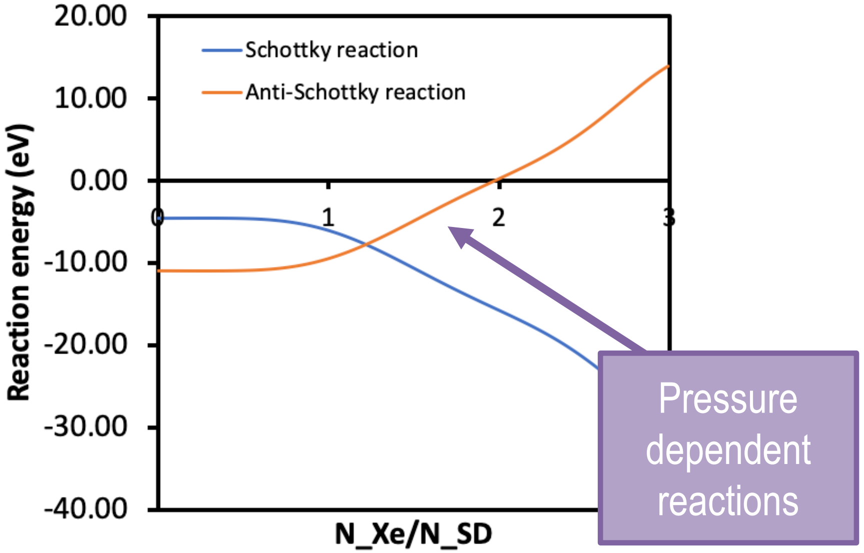


Sonoda et al. Nucl. Inst. Meth. Phys. Res. B 191 (2002) 622

Molecular and cluster dynamics of bubble pressure evolution

Defect-bubble reaction energies from MD

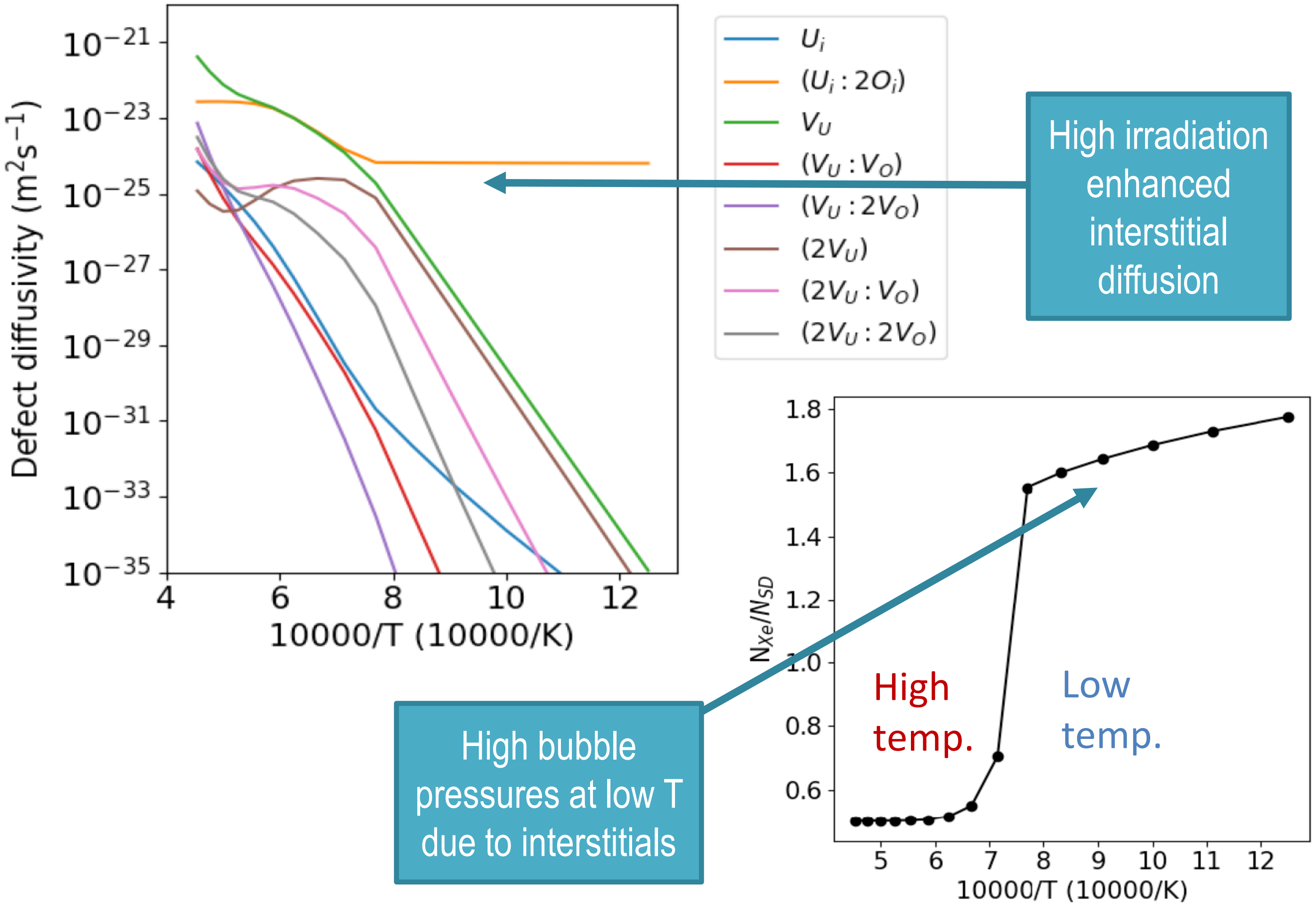




Application of ΔE_{SD} to the reaction energies for U vacancies and interstitials with bubbles

For increasing bubble pressure the reactions become more exothermic for vacancies, and less exothermic for interstitials

Bubble over-pressurization predictions from cluster dynamics



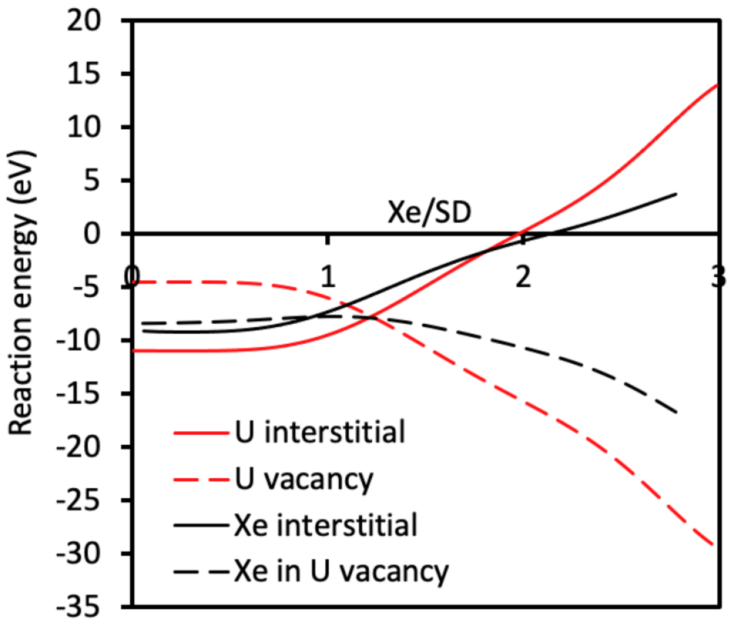
The results show that higher bubble pressures occur at the lower temperatures associated with the periphery of the pellet where HBS forms

Extension to reactions with Xe

New defect-bubble reaction energies

$$\Delta E_{Xe} = \frac{\partial(E_{bubble} - E_{void})}{\partial N_{Xe}} = 9A\left(\frac{N_{Xe}}{N_{SD}}\right)^8 + 8B\left(\frac{N_{Xe}}{N_{SD}}\right)^7 + 7C\left(\frac{N_{Xe}}{N_{SD}}\right)^6 + 6D\left(\frac{N_{Xe}}{N_{SD}}\right)^5 + 5E\left(\frac{N_{Xe}}{N_{SD}}\right)^4 + 4F\left(\frac{N_{Xe}}{N_{SD}}\right)^3 + 3G\left(\frac{N_{Xe}}{N_{SD}}\right)^2 + 2H\left(\frac{N_{Xe}}{N_{SD}}\right)$$

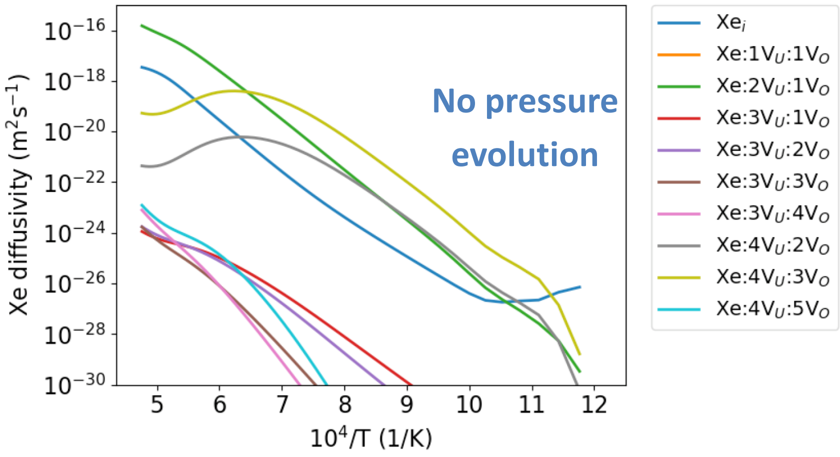
$$G^{bubble}_{\{xXe:yV_U:zU_i\}} = -G^{f}_{\{xXe:yV_U:zU_i\}} + x\Delta E_{Xe} + (y-z)\Delta E_{SD}$$

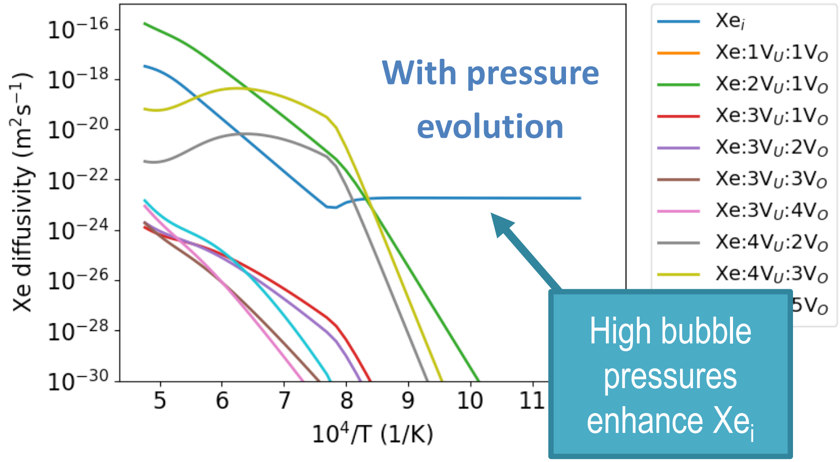


Application of ΔE_{SD} and ΔE_{Xe} to the reaction energies for Xe interstitials and Xe at vacancies with bubbles

Pressure-dependent Centipede simulations

Negligible impact on bubble pressure due to low Xe concentrations

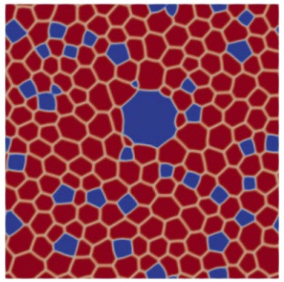




Bubble over pressurization causes Xe interstitials to play a significant role in Xe diffusion

Connection longer length scales

Expect the same point defect process to also be important for larger bubbles and are collaborating to support implementation in phase field simulations



(L Aagesen)

Explore in collaboration with INL and Pastore the addition of interstitials to Xe behavior model, so it is applicable to low T.

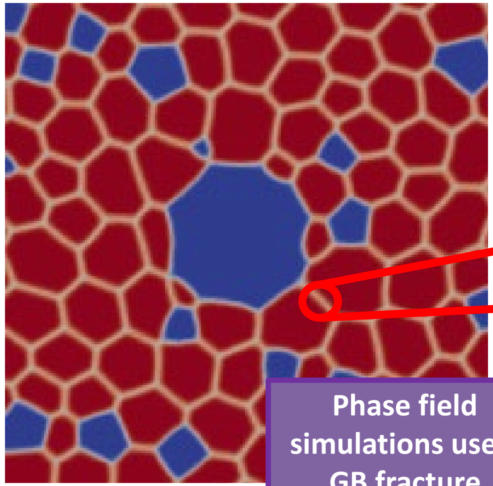
$$\frac{dn_v}{dt} = \frac{2\pi D_v \delta_g}{k_B T S}(p - p_{eq})$$ - D_i term

$$p = \frac{k_B T}{\Omega}\frac{n_g}{n_v}$$

Adjust equation of state so applicable to highly over-pressurized bubbles

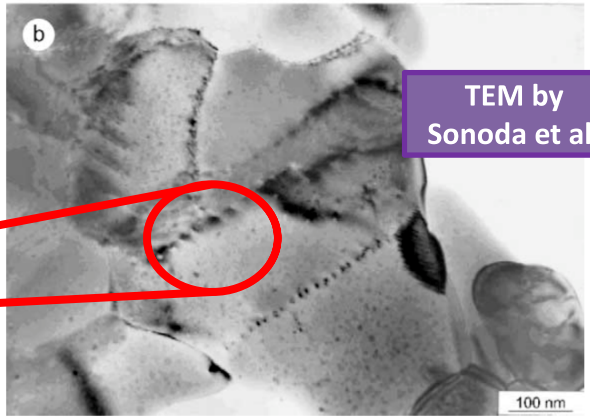
Impact of high pressure bubbles on grain boundary strength

Nm-size bubbles at sub-grains



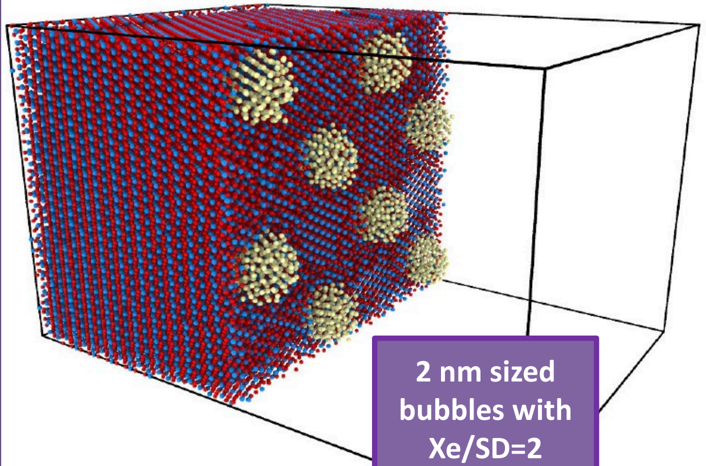
(L Aagesen)

Phase field simulations use a GB fracture criteria



TEM by Sonoda et al.

Sonoda et al. Nucl. Inst. Meth. Phys. Res. B 191 (2002) 622

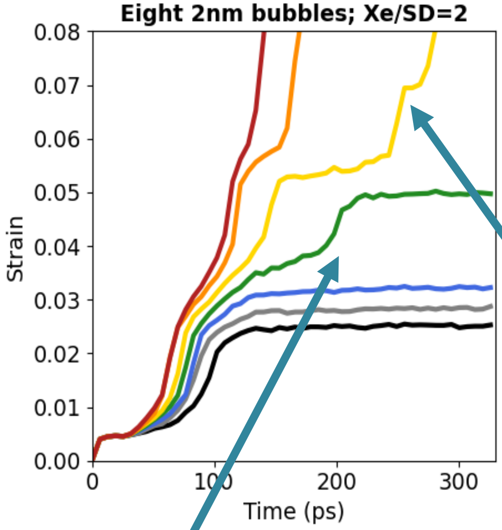


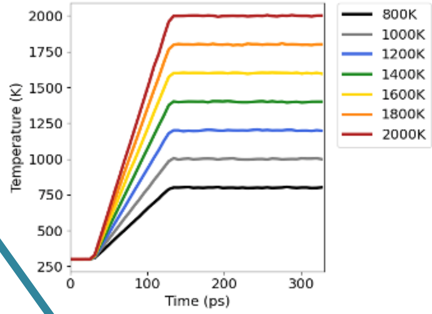
2 nm sized bubbles with Xe/SD=2

Observed small nm-sized bubbles are expected to influence the fracture strength of grain boundaries (especially if over-pressurized).

These bubbles are too small to resolve in phase field simulations, but can be examined using molecular dynamics and then the results can be used to inform more mechanistic GB fracture criteria.

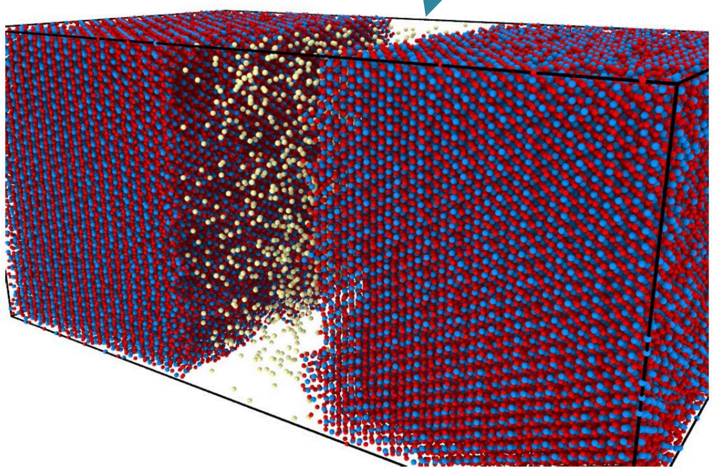
Temperature ramp with no applied stress





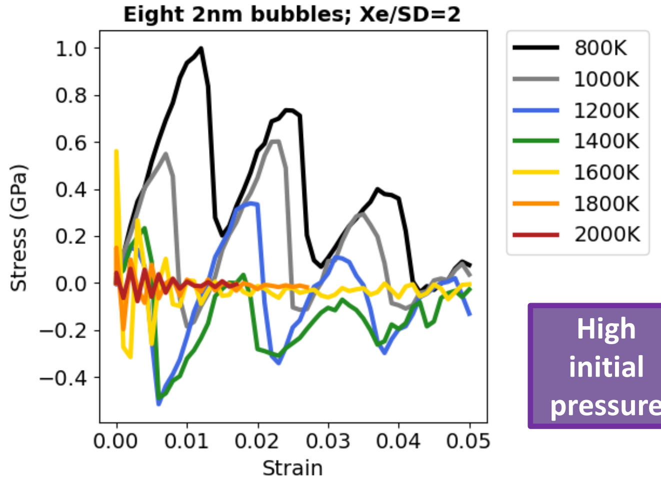
Spontaneous GB failure at high T

Evidence of micro-cracking

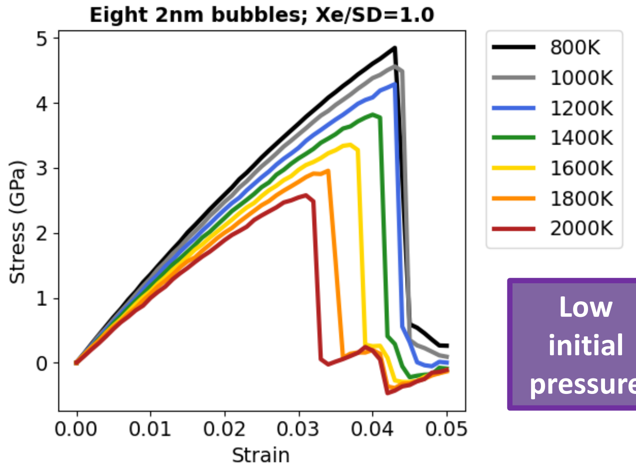


Impact of high pressure bubbles on grain boundary strength

Stress-strain analysis (impact of pressure)



High initial pressure



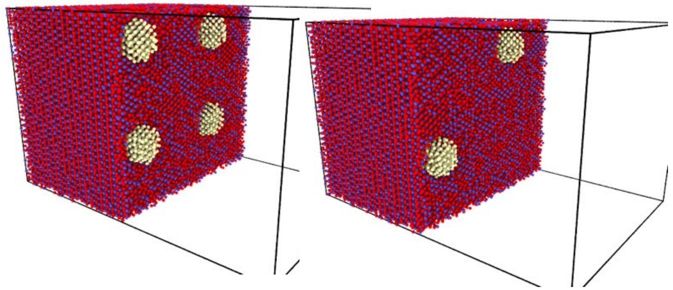
Low initial pressure

High initial bubble pressures contributes to significant grain boundary weakening.

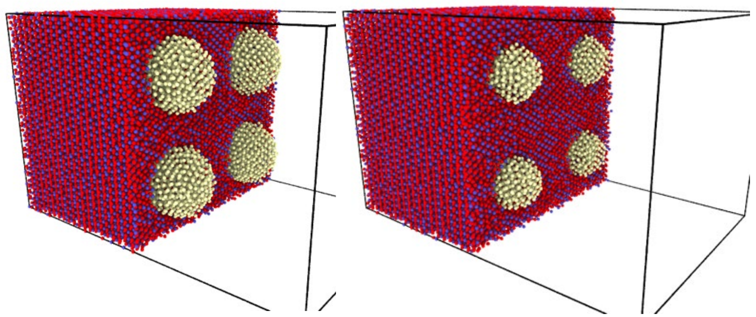
Next steps under NEAMS are to further investigate intermediate pressures (1.0-1.5).

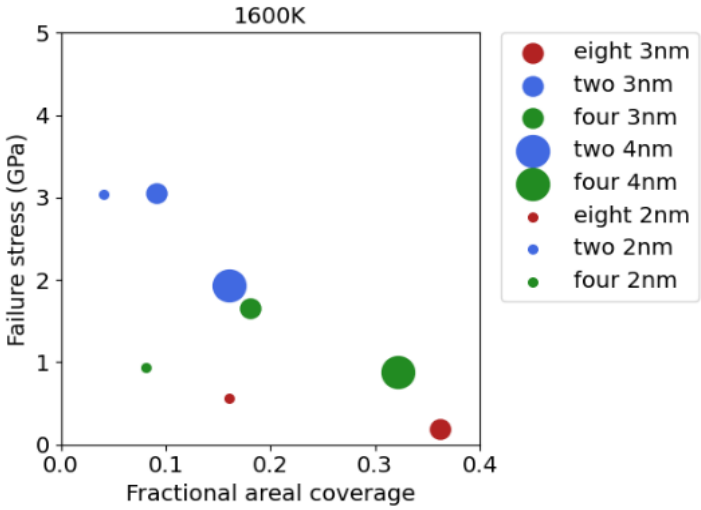
Stress-strain analysis (impact of areal coverage)

Varying number density



Varying size



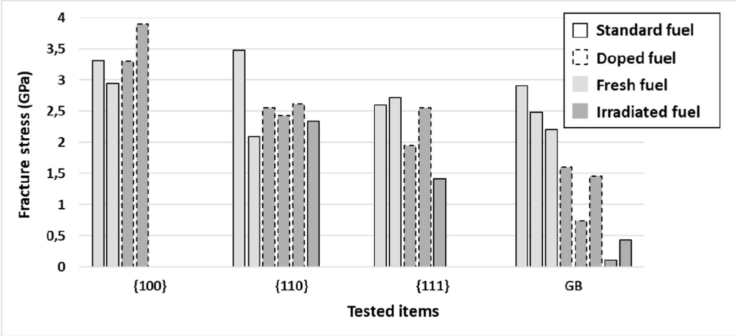


Increased areal coverage either due to larger or more numerous bubbles reduces the failure stress

Develop fracture criterion for phase field simulations of pulverization

If bubble pressure evolution can be captured in BISON pulverization criteria can be based on bubble pressure not burnup

Results will be compared to ongoing PIE micro-mechanical tests under AFC (similar to Henry et al. below)

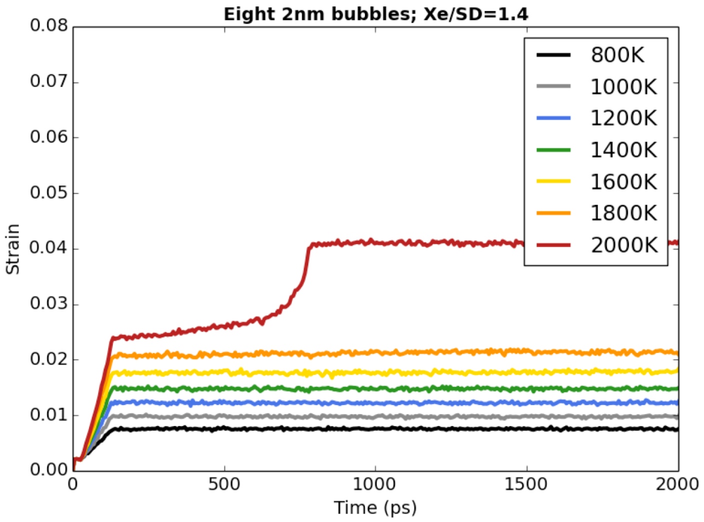


Continuation of work under NEAMS

Longer equilibration

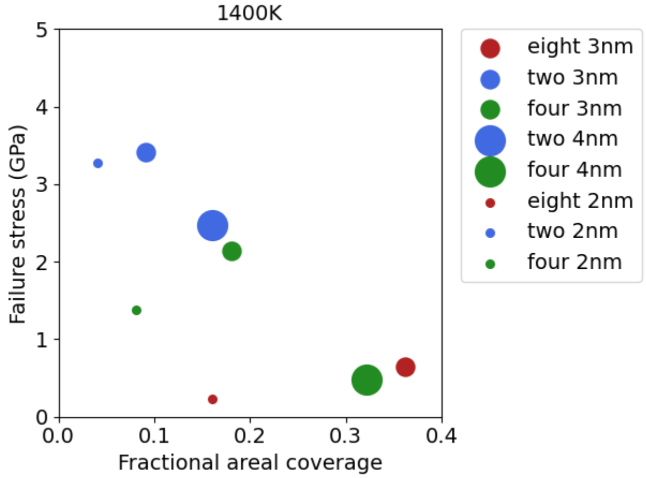
The need for long equilibration times was identified as part of the EPRI project – has been remedied under NEAMS continuation of work

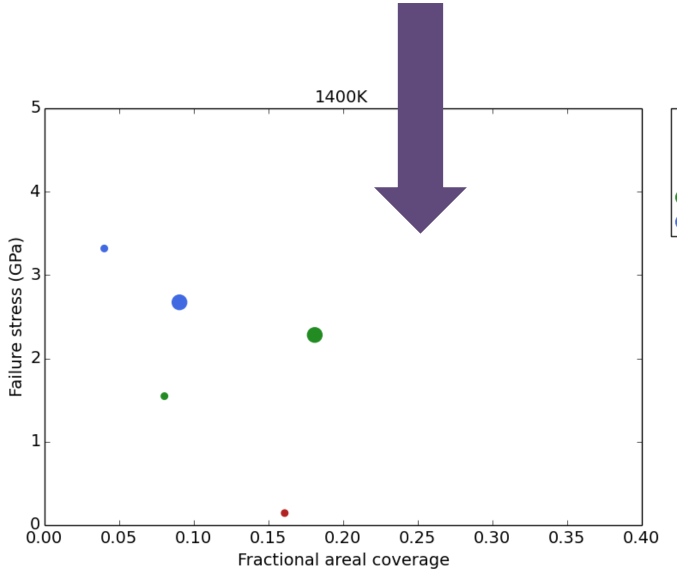
Equilibration time extended from 200 to 2000 ps



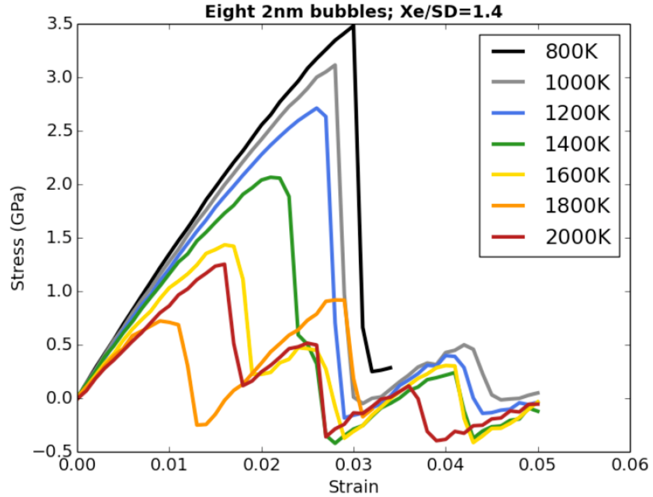
The jump in the length at 750 ps is due to a micro crack at GB causing a relaxation – highlights the need for longer equilibration times

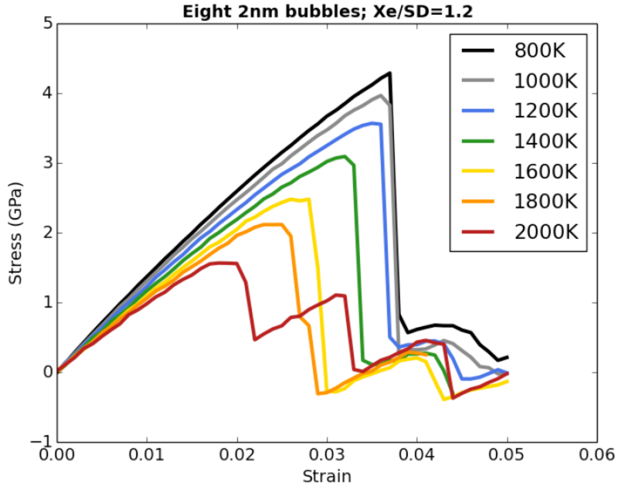
Long equilibration times have now been carried out for all cases





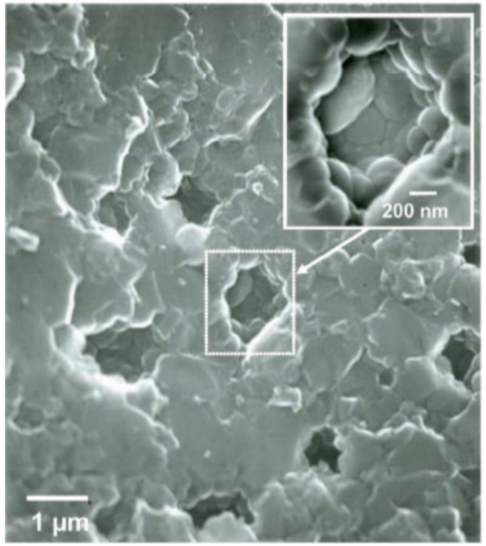
Intermediate bubble pressures

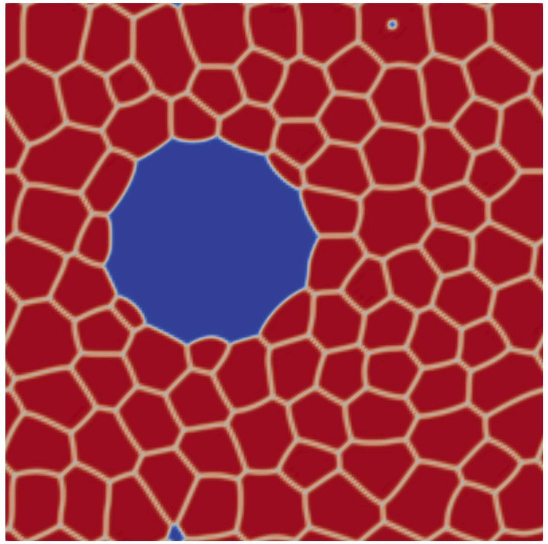




Development of physics-based pulverization criterion in BISON

- Initial focus on regions where High Burnup Structure (HBS) is fully formed
  - Grain size becomes significantly smaller
  - Bubble morphology changes from lenticular to more spherical, larger in size
- Assume pulverization occurs when pressure in a representative gas bubble in HBS region, P_g, exceeds a critical value, P_g^{cr}
  - Calculate both in BISON, compare to each other. If $P_g > P_g^{cr}$, pulverization has occurred
- Mesoscale work to inform BISON calculation of P_g, P_g^{cr}



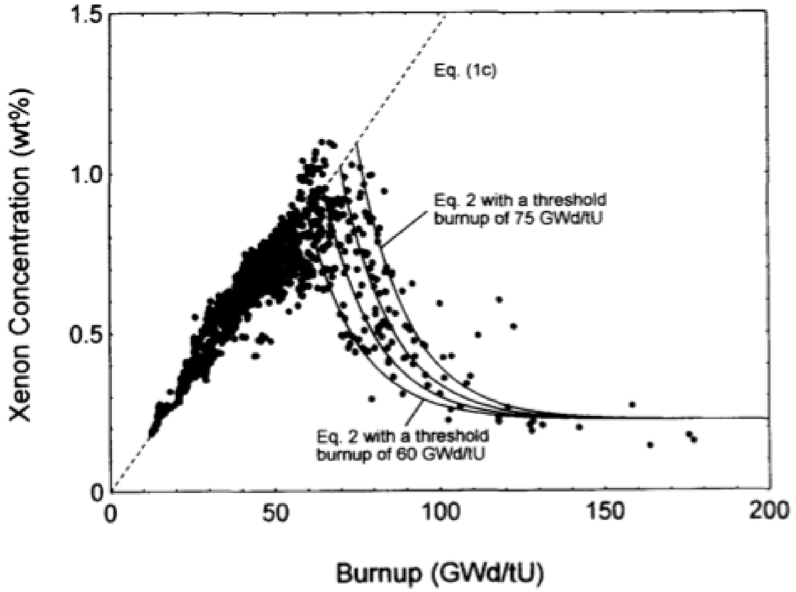


Mesoscale simulations to inform BISON calculation of Bubble Pressure

- Dislocation punching has been observed surrounding bubbles in the HBS region
  - Bubbles are likely significantly above equilibrium pressure for their size (Nogita & Une, JNM, 226, p. 302, 1995)
- Hypothesized mechanism of overpressurization:
  - Intragranular Xe peaks prior to HBS formation
  - When HBS forms, many new subgrain boundaries form. These allow a path for intragranular Xe to be transported to existing bubbles, increasing pressure and causing growth
- Pressure could peak much higher than dislocation punching pressure, then be relieved through growth.
  - How much higher?
  - Can we use dislocation punching pressure as an estimate for initial P_g?

Dislocation punching pressure:

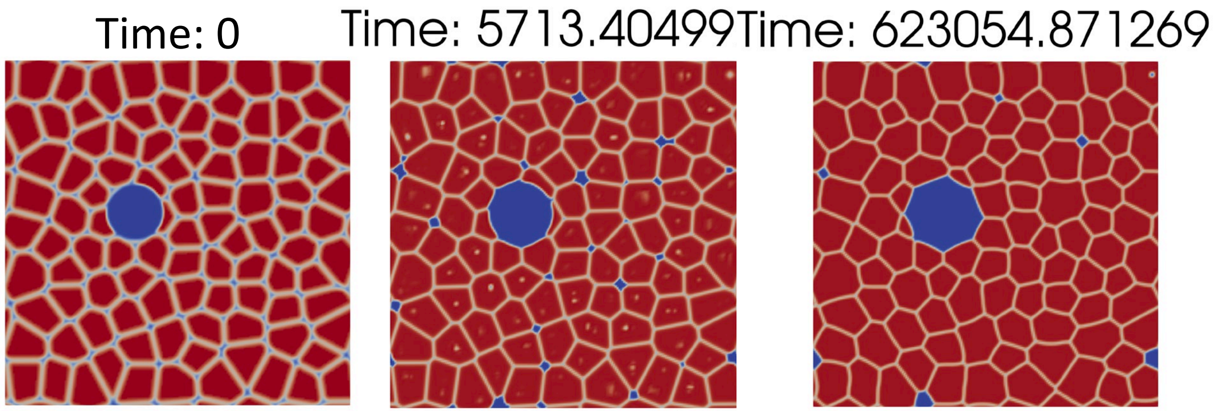
$$P = P_{eq} + P_{ex} = P_H + \frac{2\gamma}{r} + \frac{Gb}{r}$$



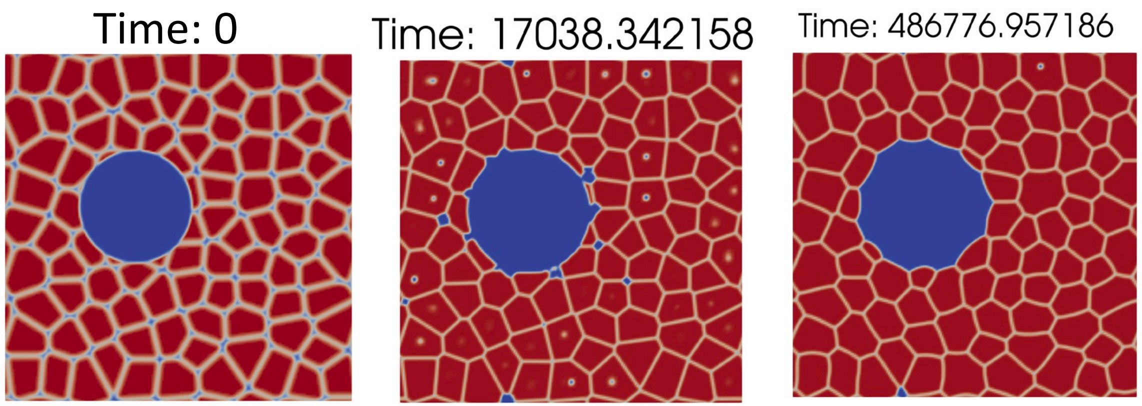
Intragranular Xe concentration
Lassman et al., JNM (1995)

Mesoscale simulations to inform BISON calculation of Bubble Pressure

- Simulate growth of bubbles just after onset of HBS formation with phase-field modeling:
  - Order parameters to represent the bubble phase and multiple grains of matrix phase
  - Defect species concentrations: vacancies and Xe atoms
  - Bulk defect diffusivities and steady-state defect concentrations parameterized from atomistic/cluster dynamics simulations (LANL). Initial bubble pressures at equilibrium
  - Enhanced diffusivities along grain boundaries and bubble-matrix interface
  - Matrix supersaturated with Xe in initial conditions based on peak value of experimental data
  - Calculate bubble pressure during growth

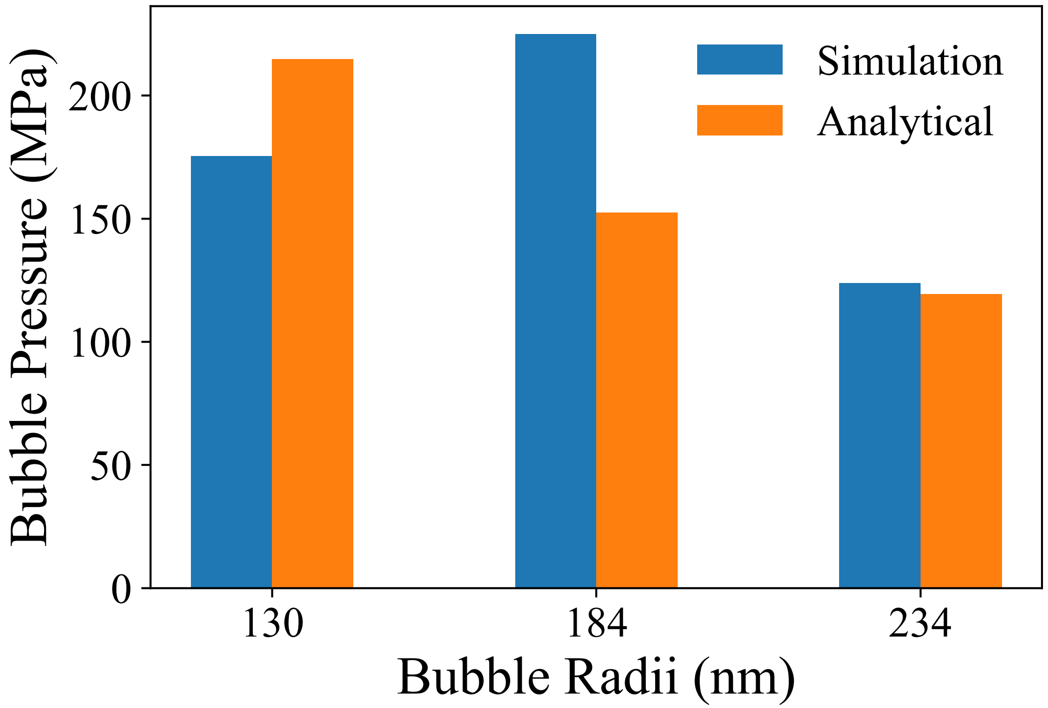


Initial bubble radius: 100 nm



Initial bubble radius: 200 nm

Peak Pressures from Mesoscale Simulations and Implications for BISON



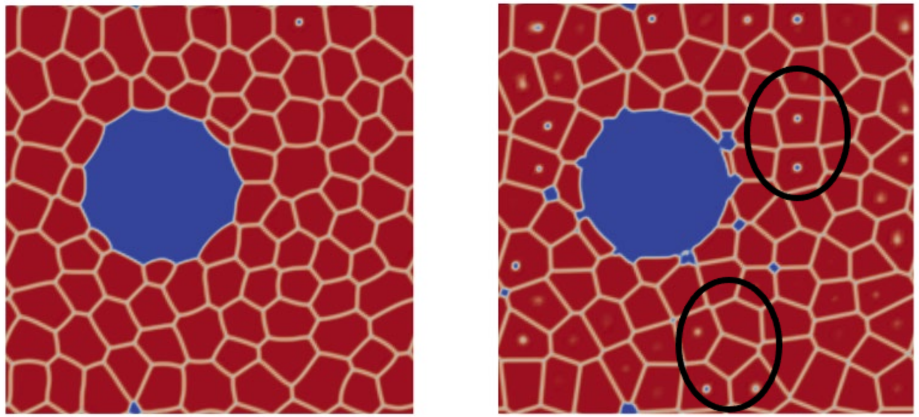
Comparison of peak bubble pressure from Simulation to Analytical dislocation punching pressure:

$$P = P_{eq} + P_{ex} = P_H + \frac{2\gamma}{r} + \frac{Gb}{r}$$

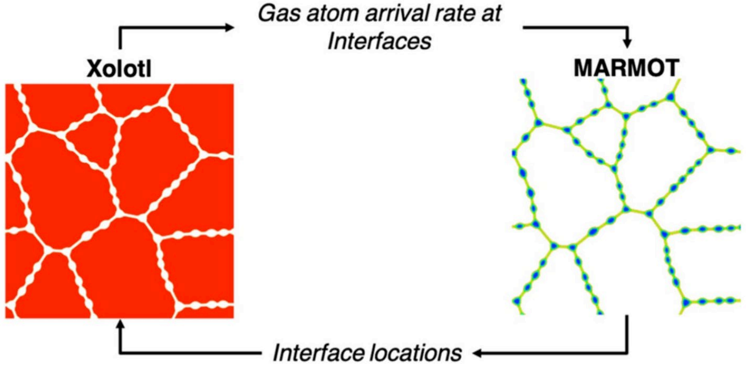
- For bubble sizes considered, peak pressure is comparable to dislocation punching pressure
  - **Based on this, use dislocation punching pressure for estimate of initial pressure of HBS bubbles for BISON**
- Bubbles are still significantly overpressurized relative to equilibrium
  - **Evolution equation for bubble pressure following HBS formation is still needed**

Marmot-Xolotl Coupling Approach

- Morphology predicted by the model matches experimental observations
- Intragranular bubbles form instantaneously within the grains, not tractable at mesoscale
- Xolotl coupling to separate out the intergranular and intragranular Xe atoms



Intragranular Bubble Formation



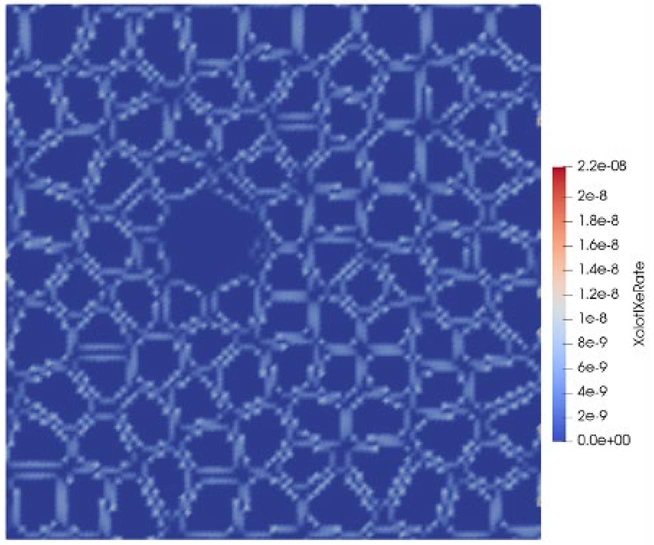
The Coupled Approach (Kim et al., Mat. Th., 2021)

Modified Approach for HBS

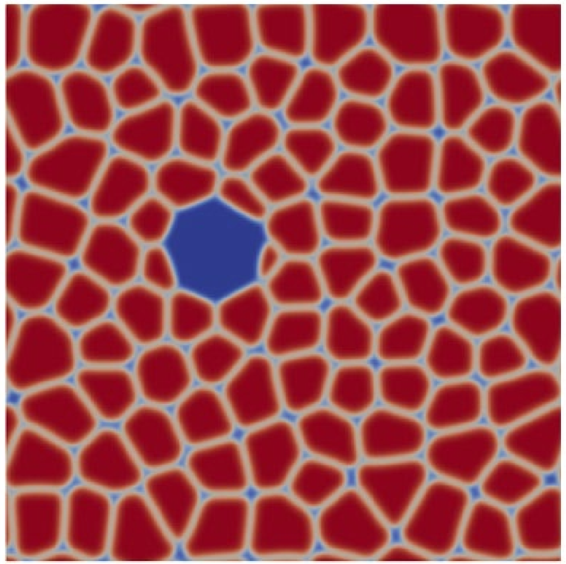
1. Determine the time till HBS initiation
2. Run 0D Xolotl simulations to determine the Xe evolution before HBS formation
3. Generate spatial distribution of Xe and use it as initial condition for 2D Xolotl simulations
4. Transfer Xe concentration to phase-field model, initialize the microstructure
5. Concurrently solve both the models using MultiApp

Marmot-Xolotl Coupling Results

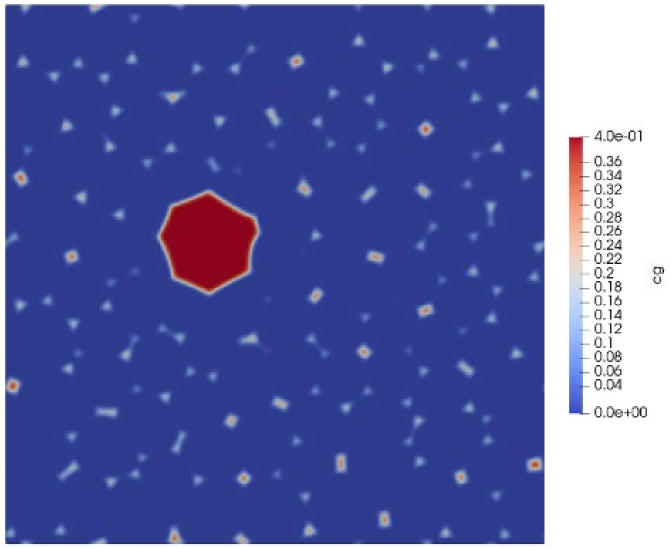
- 1.25 µm X 1.25 µm domain with 100 grains and single bubble
- The 2D Xolotl simulations include diffusion, clustering, and re-solution
- Additional single Xe atoms released due to diffusion and re-solution is added as source term at the grain boundaries
- Vacancy source term is proportional to the Xe source
- The numerical issue is resolved, the model captures realistic behavior. **Use this improved approach to determine initial pressure of HBS bubbles.**



Xe Source



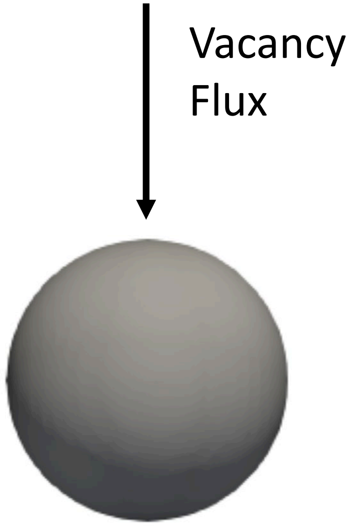
Microstructural Evolution



Xe Concentration

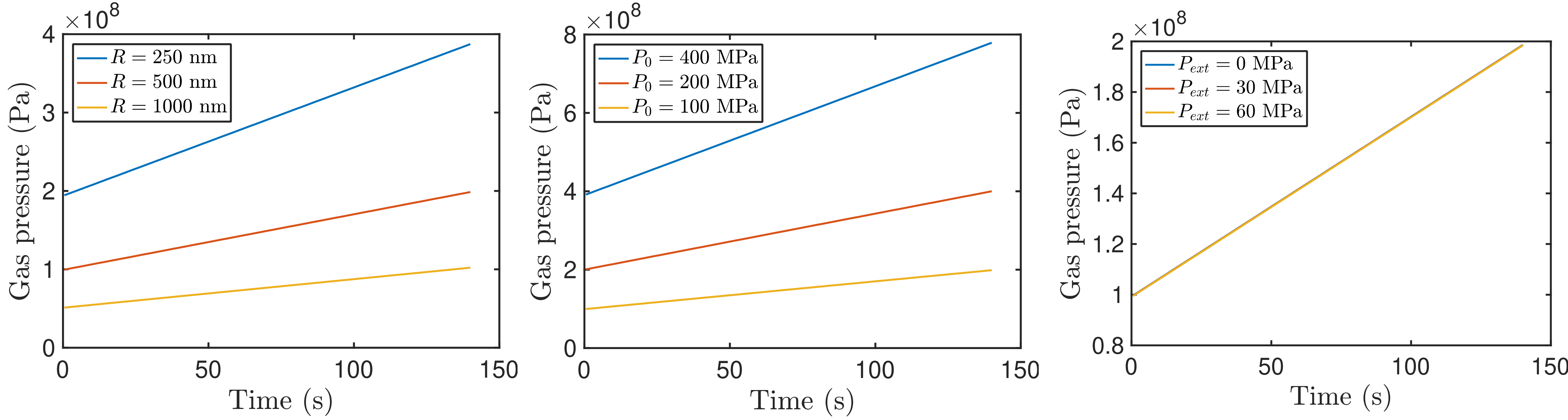
HBS bubble response to LOCA transient

- 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?**
  - **What is the pressure response to a given temperature transient?**

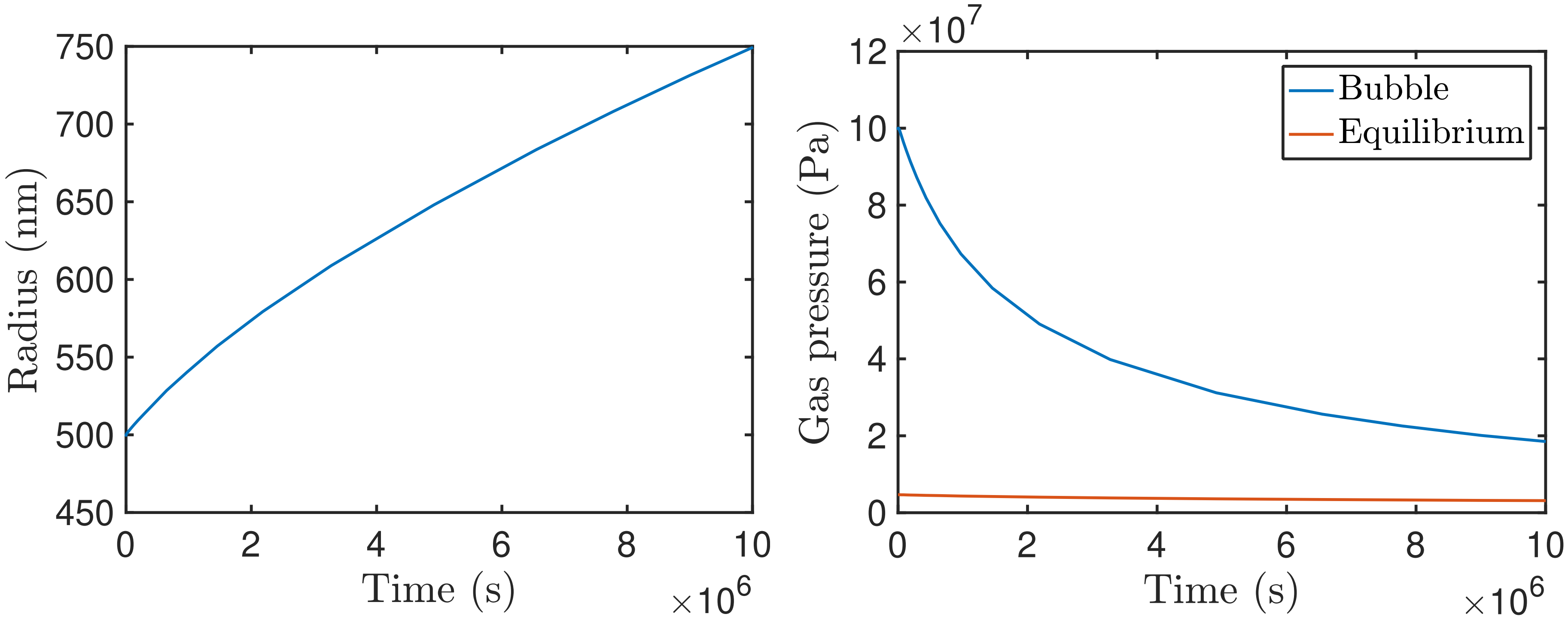


HBS Bubble response to LOCA transient

- Developed of new phase-field model to address LOCA behavior:
  - Includes surface tension of bubble-matrix interface and gas pressure; allows consideration of effect of overpressurized bubbles
- LOCA transients: variations in bubble size, initial pressure, porosity, external pressure
- **Bubble size does not change significantly for any cases considered**
- **Pressure as a function of time passed to PF fracture model**



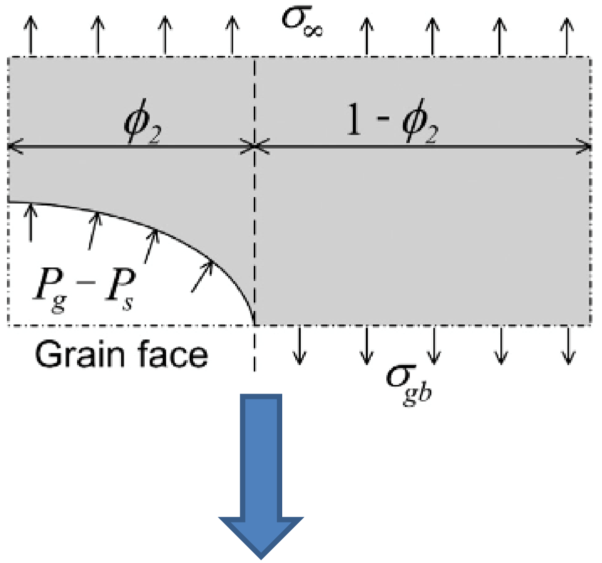
Bubble growth during steady-state operation



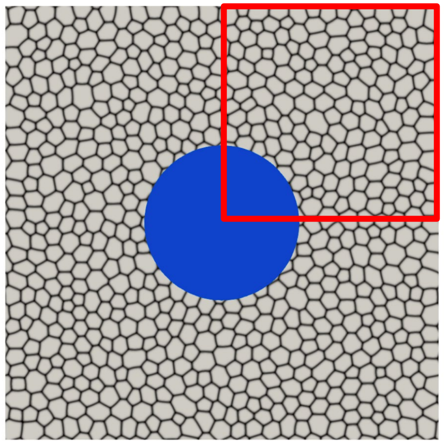
- Assume bubble pressure is 100 MPa in initial conditions
  - Upper bound based on dislocation punching pressure
- Bubble pressure decreases during growth but remains well above equilibrium pressure
  - Increased likelihood of fragmentation during LOCA

Development of Physics-Based Analytical Criterion for Pulverization

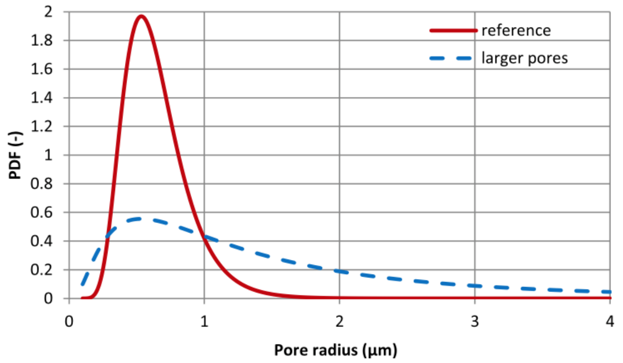
- Began implementation of pulverization based on analytical expression (Olander)
  - To be complemented by Phase-Field Fracture results
- Originally developed for lenticular (non-HBS) bubbles
  - Adapted to HBS geometry using porosity, assume worst-case scenario of flat GB
- Pulverization occurs when $P_g > P_g^{cr}$
- Determine P_g during transient for most frequently occurring bubble, R = 0.53 μm
  - Starting from dislocation punching pressure



$$P_g^{cr} = P_s + \frac{\sigma_{gb}^{cr}(1-\phi_2) - \sigma_\infty}{\phi_2}.$$



Determine ϕ_2 from porosity (empirical)



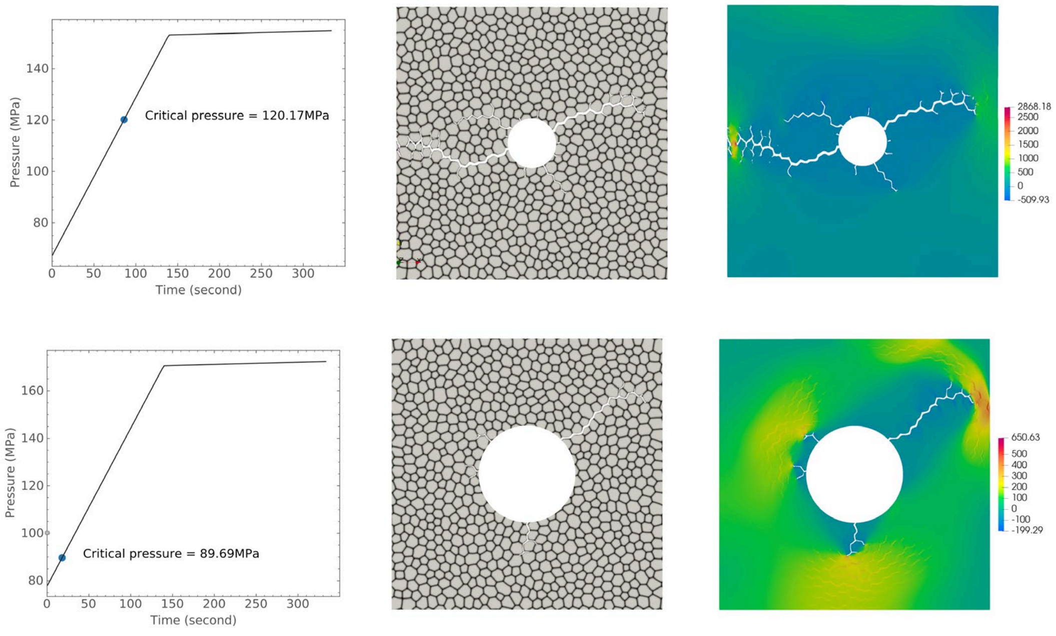
Kulacsy, JNM 466, 409-416 (2015)

Phase-field fragmentation modeling

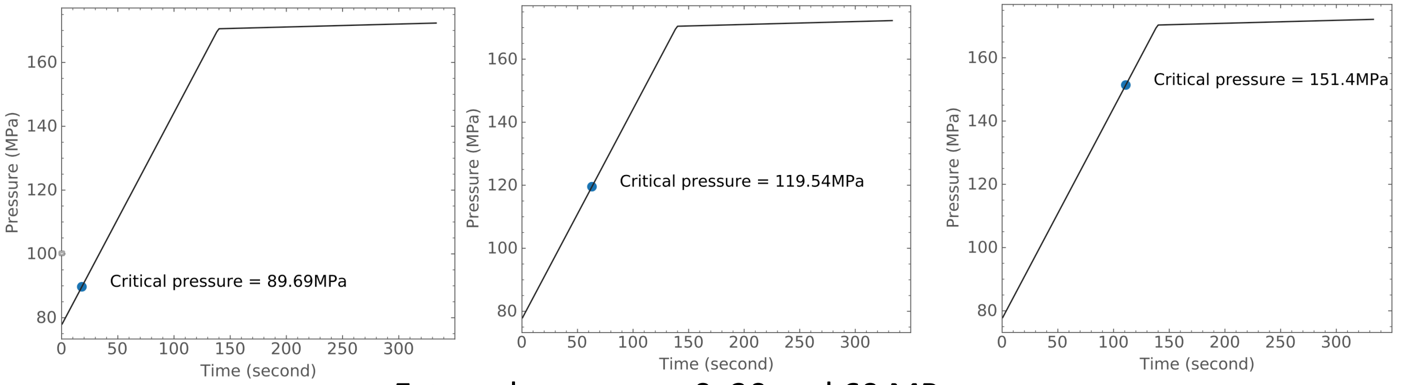
Developed a phase-field model for quasi-brittle pressurized fracture

Phase-field fracture model was used to study HBS fragmentation behaviors

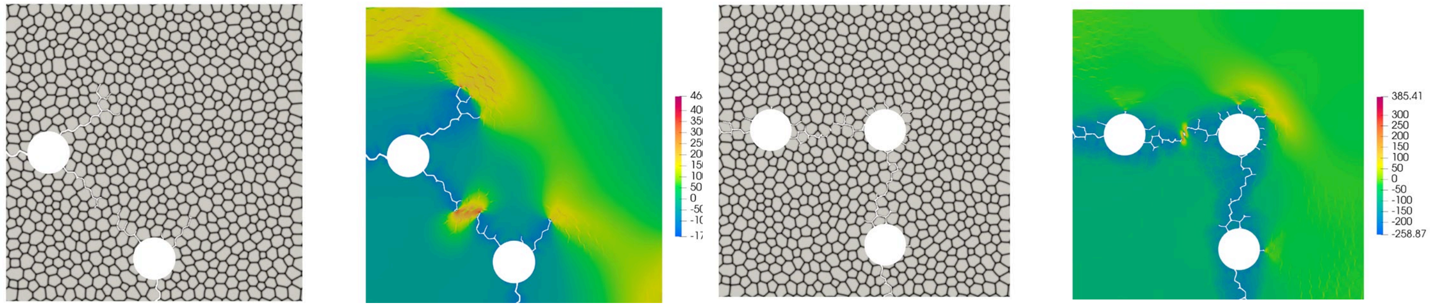
- Effect of bubble sizes: critical pressure is lower for the larger bubble
- Effect of external pressure: critical pressure becomes higher for larger external pressure values
- Effect of bubble interaction: fragmentation size is likely determined by bubble spatial distribution



Bubble size = 0.25 μm and 0.5 μm



External pressure = 0, 30 and 60 MPa

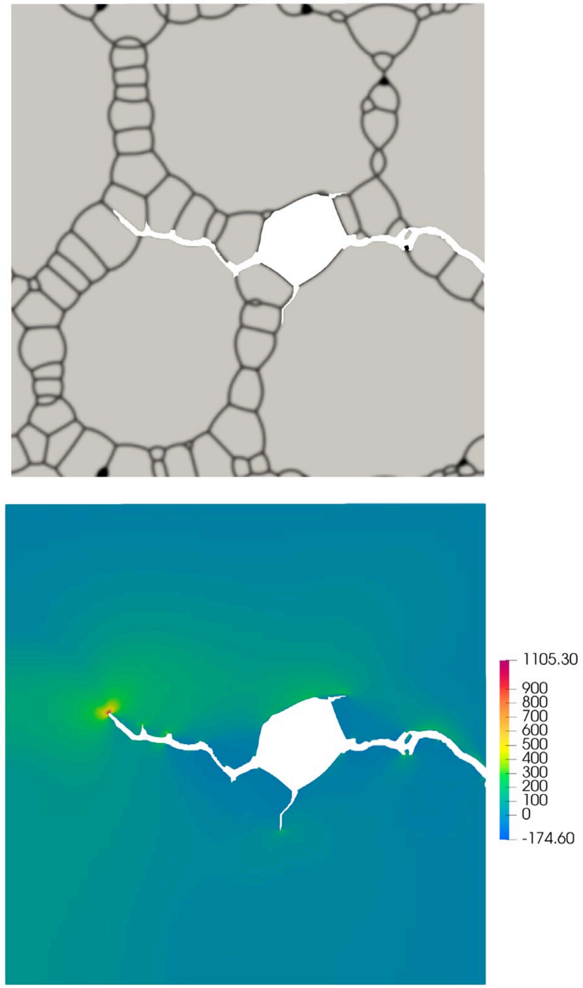


Two and three bubbles interaction

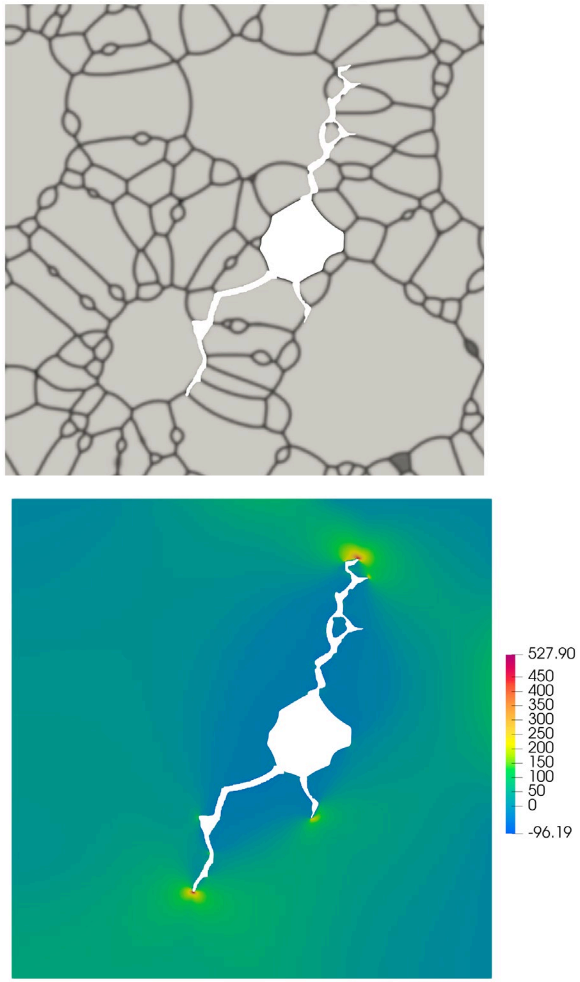
Fragmentation in partial HBS

Phase-field fracture model was used to simulate fragmentation behaviors of partial HBS

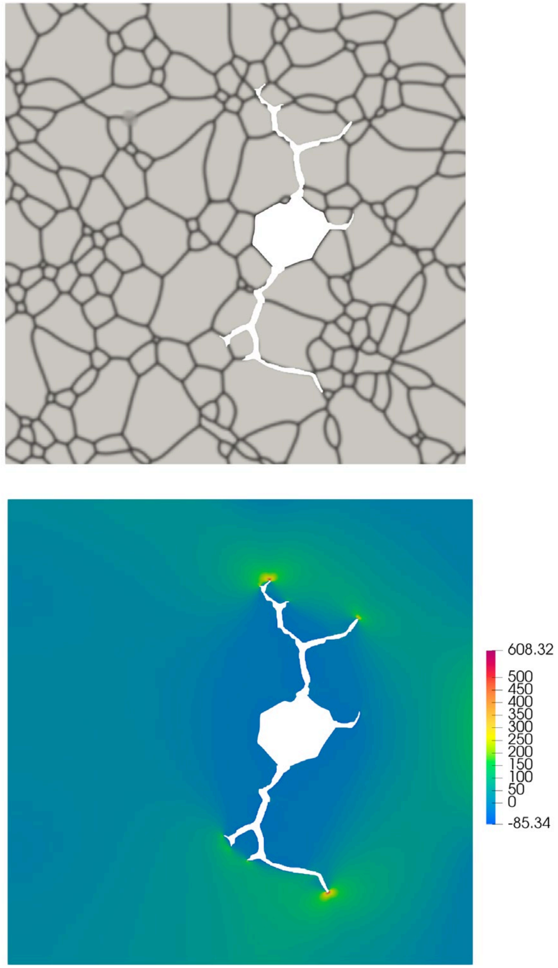
- Use the output from the HBS formation simulations as our initial condition
- Three HBS at different recrystallization stages with 25%, 60% and 100% recrystallization fraction were considered
- Crack initiation locations and crack propagation paths varied among the three cases because recrystallized grain structures change.



25 % recrystallization stage



60 % recrystallization stage

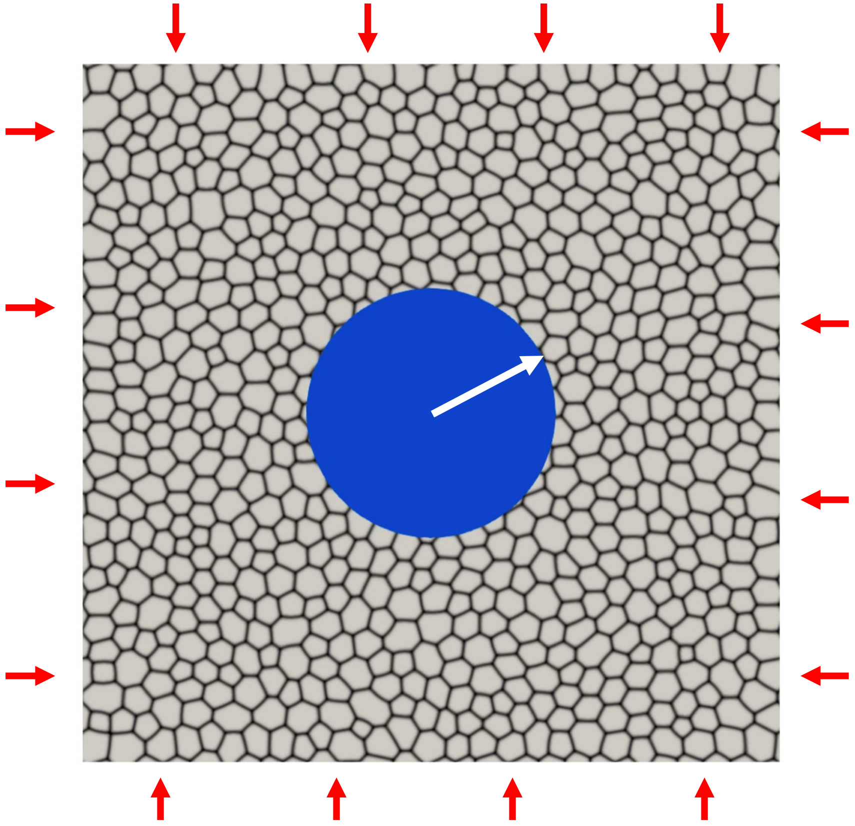


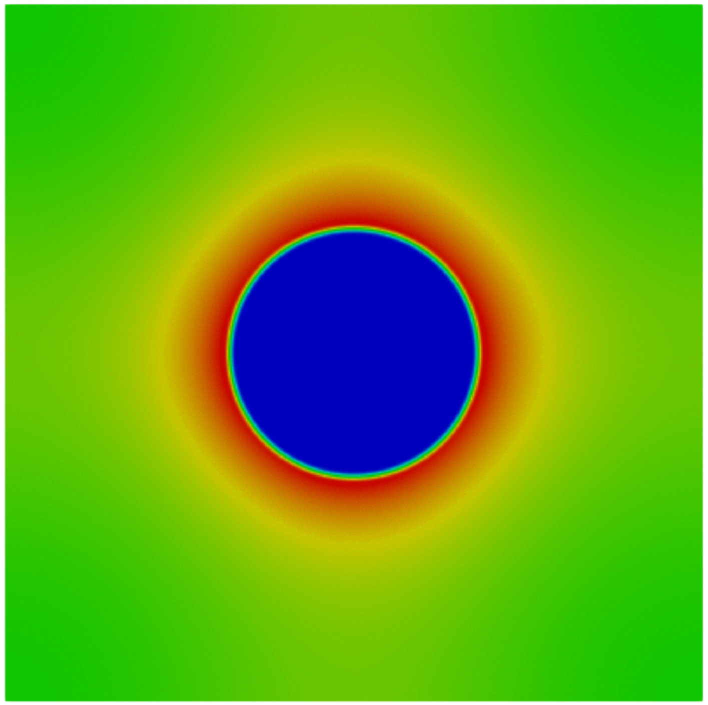
100 % recrystallization stage

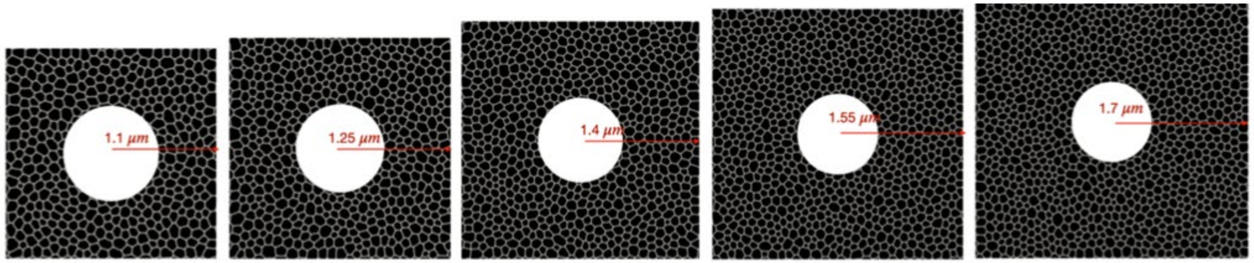
Determine BISON pulverization criterion based on phase-field modeling

Phase-field fracture model was used to inform pulverization criterion for BISON models

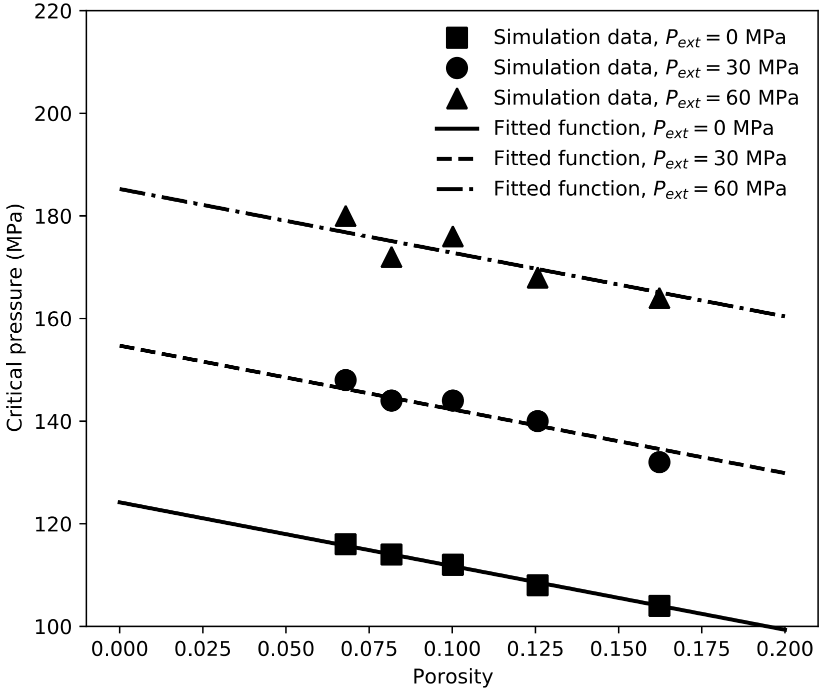
- Use periodic boundary conditions to account for multi-bubble interaction.
- Consider varying porosity (p), external pressure (p_{ext}), critical fracture stress of grain boundary (σ_c)

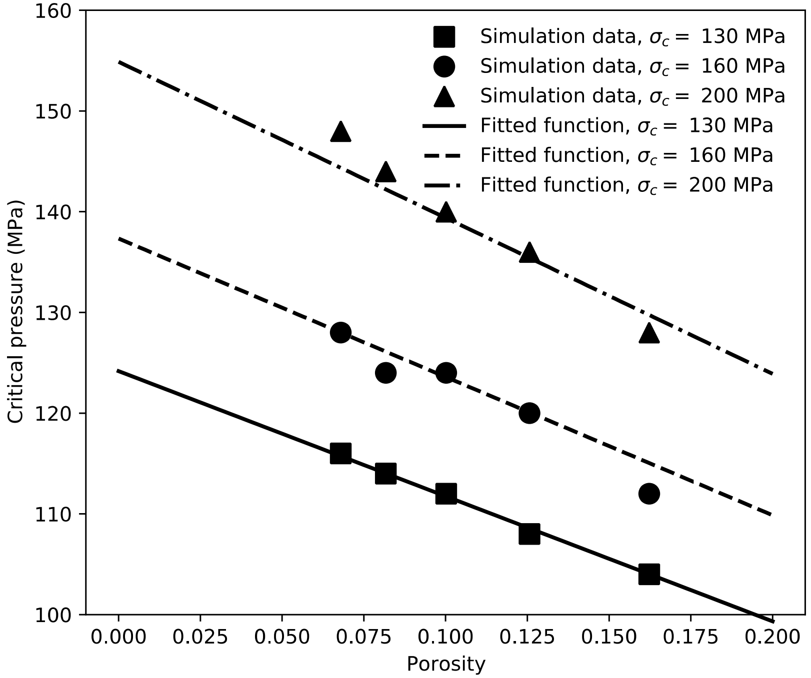






BISON pulverization criterion





- Function to fit the data (a, b, c are fitting constants) :

$$P_g^{cr} = [a + b(\sigma_{cr} - 130)](1 - p) - cP_{ext}$$

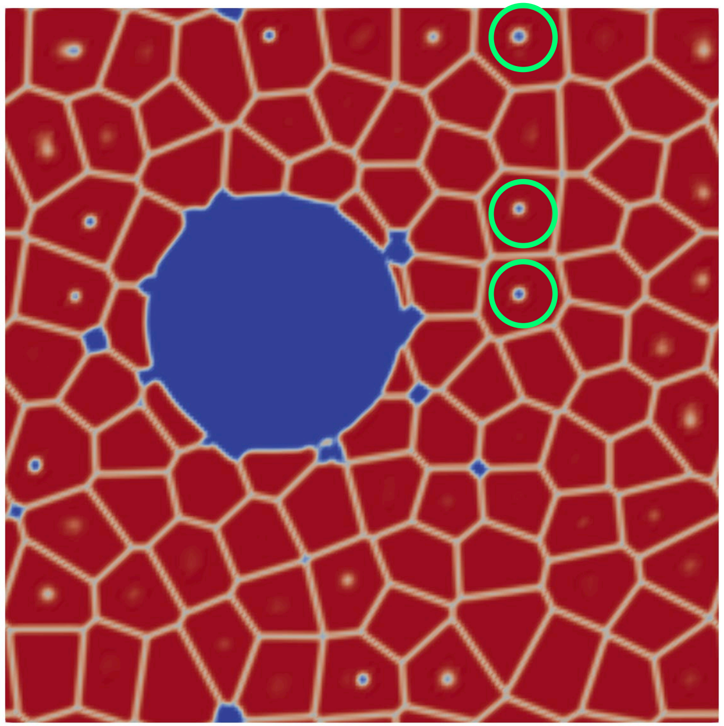
- Implemented in BISON. Validation with BISON assessment cases is in progress, using both analytical and phase-field fracture-based criteria
- Future work:
  - Inform σ_c, G_c from atomistic simulations (LANL) and experiment (AFC)

Conclusions

- Atomistic:
  - Nano-meter size bubbles at grain boundaries have high pressure due to radiation-enhanced interstitial diffusivity
  - These same small bubbles significantly affect strength of grain boundaries. Work in progress to further quantify effect to inform mesoscale fracture model
- Mesoscale:
  - Phase-field simulations support using dislocation punching pressure as initial pressure of HBS bubbles
  - Bubble size does not change significantly during the duration of a LOCA transient, simplifying the requirements on the fracture model
  - Fracture model was developed to predict when pulverization will occur. BISON model developed based on simplify geometry, 2D. Future work: extend to 3D, consider multi-bubble interactions, partial HBS regions
- Engineering scale: initial validation done with BISON model (K. Gamble talk)

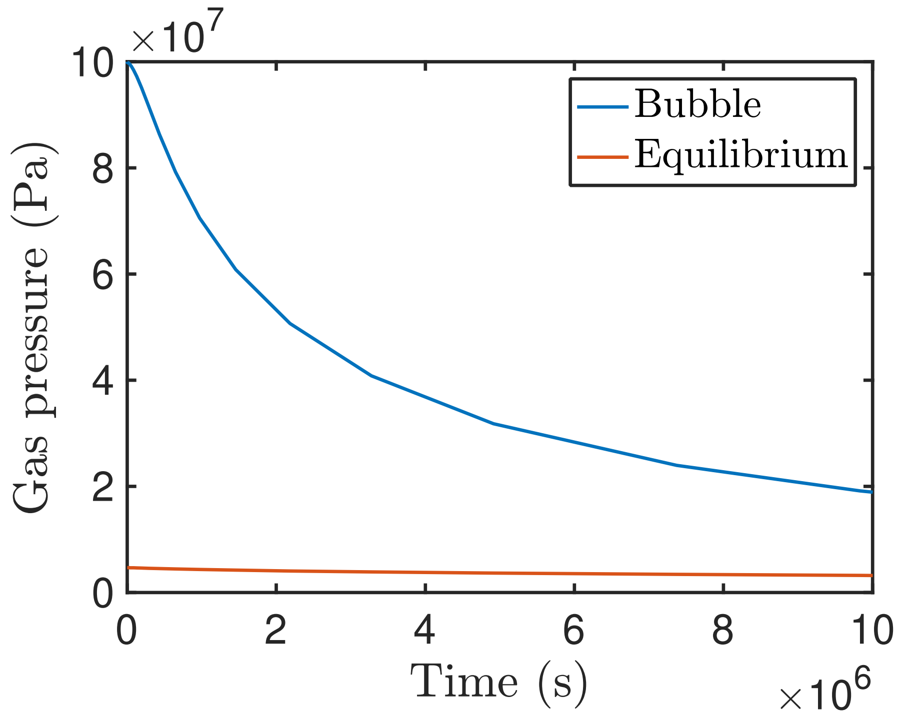
FY22 plans

- Improve estimates of initial HBS bubble pressure at formation:
  - High gas supersaturation in initial conditions results in intragranular bubble formation
  - Phase-field model length scale not small enough to resolve intragranular bubbles at realistic size
  - Use coupled Marmot-Xolotl approach developed by SciDAC project
  - Current phase-field model assumes monovacancies and Xe on monovacancy sites
  - Add (minimum number of) defect species and reactions to better capture transport at the mesoscale



FY22 plans

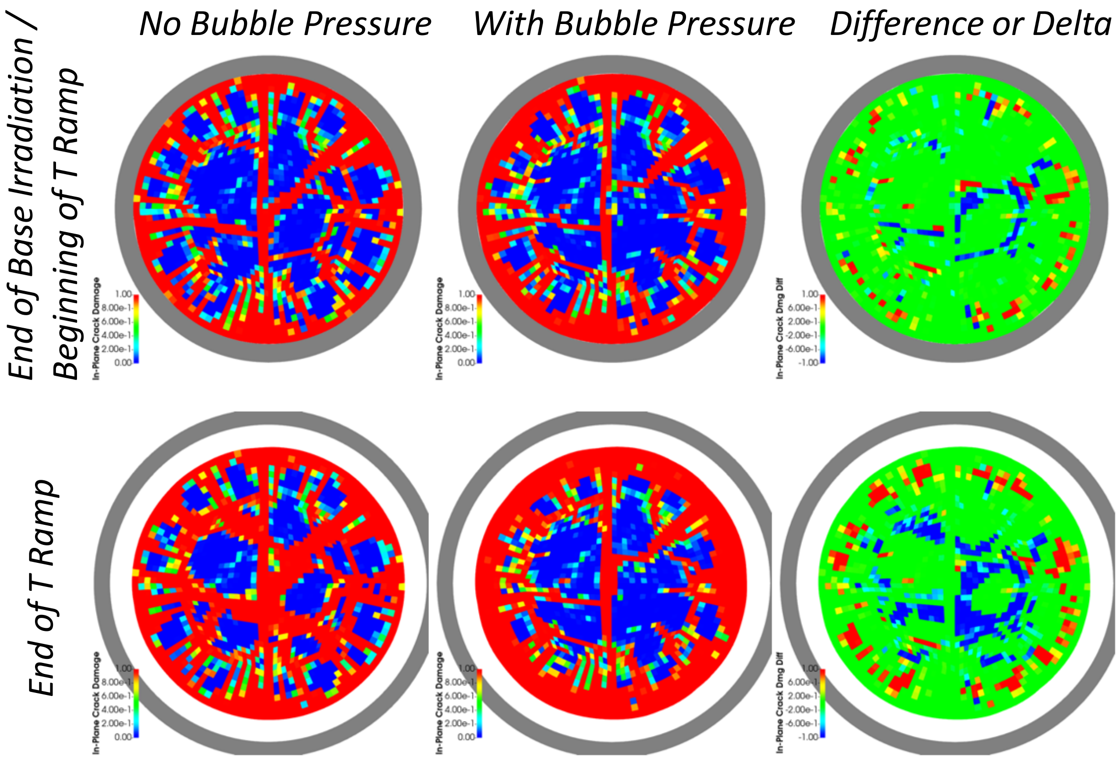
- Model for HBS bubble pressure evolution (after formation)
  - Based on current phase-field results, assume growth driven by vacancy flux
  - KKS phase-field model: even starting at dislocation punching pressure, bubble pressure remains considerably above equilibrium
  - Goal: Analytical model
- Improve fragmentation criterion:
  - Incorporate σ_c, G_c from atomistic simulations (LANL) and experiment (AFC)
  - 3D simulations



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

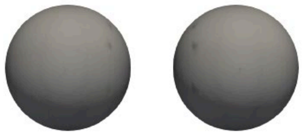
FY22 plans

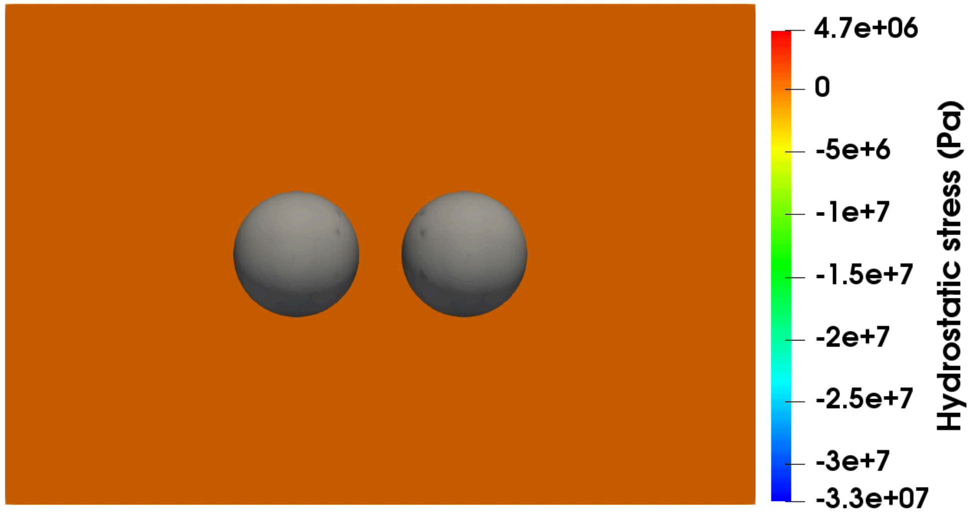
- Include effects of pore pressurization and pulverization on engineering-scale response
  - EPRI-funded study in FY21 showed how fragmentation driven by pore pressurization can be captured using an effective stress approach and smeared cracking model in Bison.
  - This did not account for effects of burnup on strength (which are now captured in pulverization criterion)
  - Update this approach to use pulverization criterion together with cracking model, which will account for stress redistribution due to pulverization



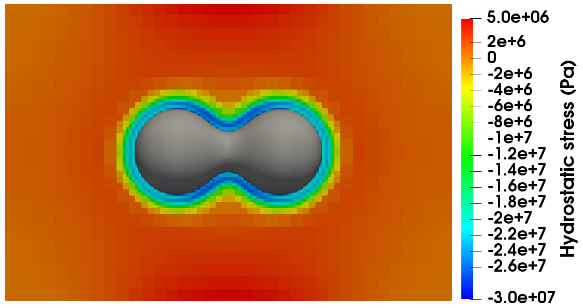
*Simulation of Fracture Damage (extent of fuel fragmentation) for **Rodlet No. 191** from NRC-sponsored semi-integral LOCA tests with and without the effect of bubble pressure (refer to NUREG-2121) using Bison with pore pressurization and smeared cracking models.*

Bubble growth during steady-state operation





- 3D simulation to 1.5×10^7 s, 2 bubble impingement, initial radii of 300 nm
- Hydrostatic stress surrounding bubbles
  - Region of enhanced compressive hydrostatic stress in “neck”



$t = 1.3 \times 10^6$ s