INL/MIS-21-63511-Revision-0

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July 2021

Changing the World's Energy Future

Larry K Aagesen Jr, Stephen R Novascone, David Andersson, Wen Jiang, Sudipta Biswas, Michael Cooper, Christopher Matthews, Nathan Capps

Idaho National Laboratory

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**Idaho Falls, Idaho 83415**

**http://www.inl.gov**

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# NEAMS Development of a Mechanistic High Burnup $UO_2$ Pulverization Model for Engineering Application

**Fuels Leads:** Stephen Novascone[1] and David Andersson[2]

**Technical Team:** Larry Aagesen[1], Wen Jiang[1],Sudipta Biswas[1], Michael Cooper[2], Topher Matthews[2], and Nathan Capps[3]

[1]Idaho National Laboratory
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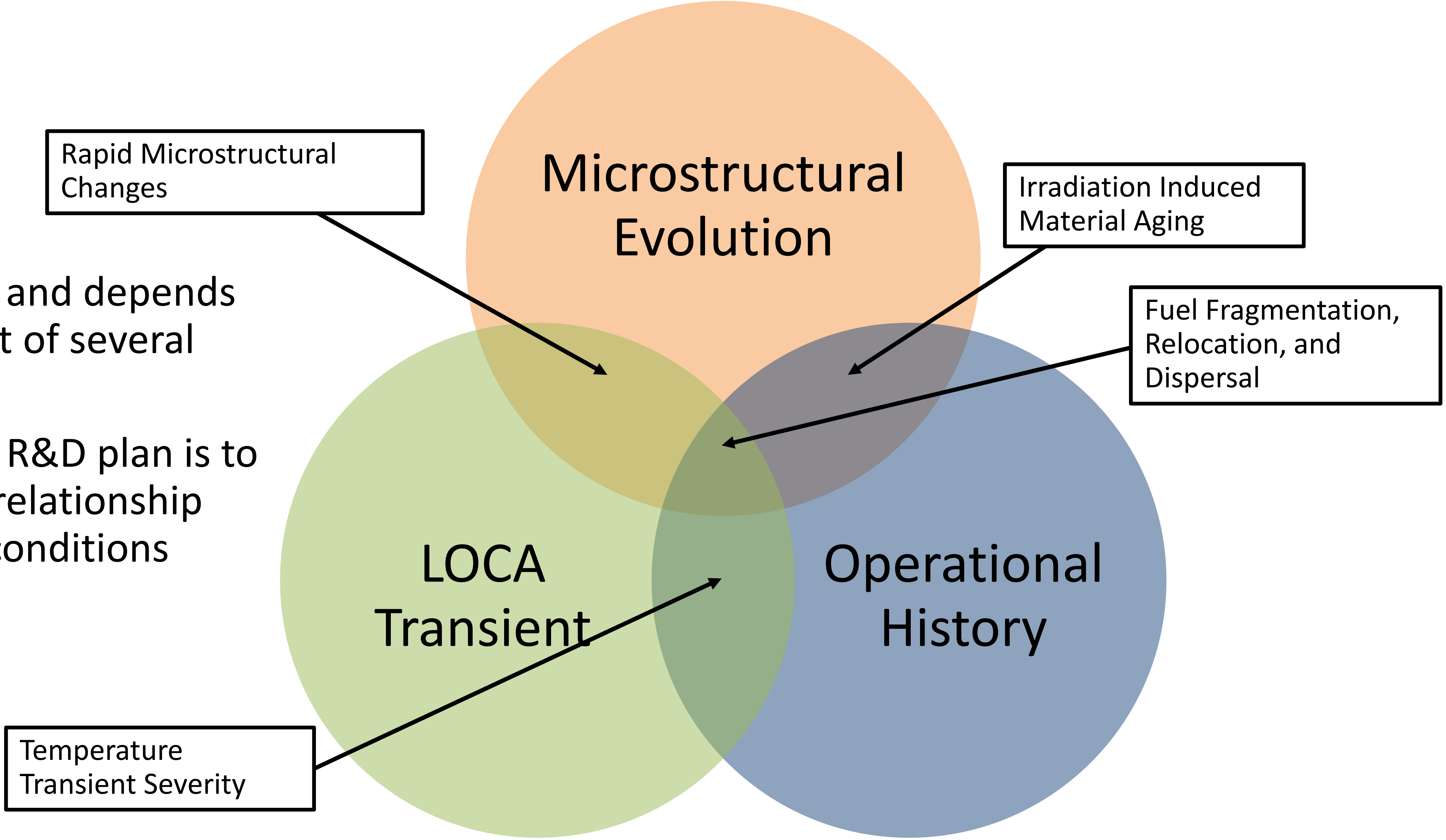
[3]Oak Ridge National Laboratory

U.S. DEPARTMENT OF ENERGY

# Mechanisms Driving FFRD

- FFRD is complex and depends on the alignment of several conditions
- The focus of our R&D plan is to understand the relationship between these conditions



# High Burnup $UO_2$ Pulverization Model

### Atomic Scale Mechanisms

- Steady State Bubble Pressure – cluster dynamics
  - Determines irradiation-enhanced defect processes to be included in meso-scale
  - Predicts bubble pressure for use in MD
- Transient GB Fracture – MD simulations
  - Informs meso-scale with GB failure criteria
  - Captures role of high pressure GB bubbles

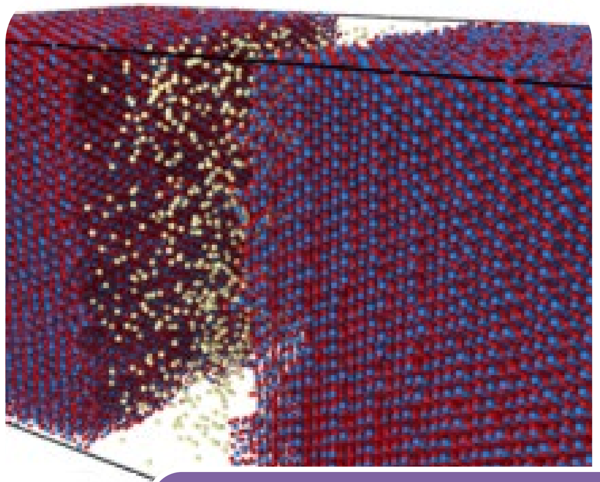
### High Fidelity Model Development

- HBu microstructure formation
- FG bubble nucleation and growth
- Microstructure mechanical degradation
- Inform transient FGR (temperature transient)

High fidelity models to inform engineering scale high burnup $UO_2$ pulverization model

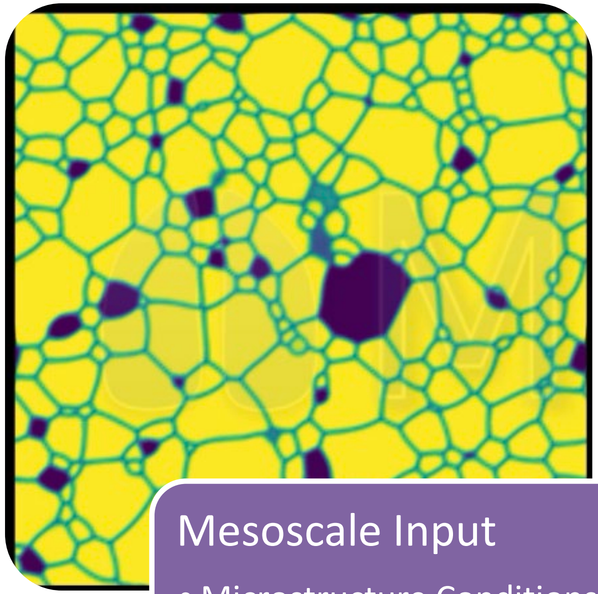
### Engineering Scale Application

- Steady State
  - Define condition of the microstructure as a function local pellet conditions (i.e., LHR, burnup, temperature, radius, etc.)
- Transient
  - Predict pulverization as a function of pellet radius and thickness of pulverized region



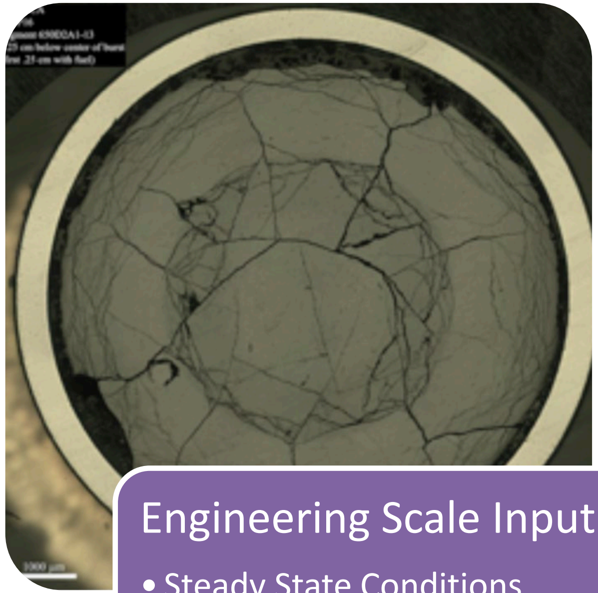
### Atomistic Input

- FG bubble pressures
  - Steady state and transient
- Local grain boundary mechanical properties



### Mesoscale Input

- Microstructure Conditions
- Steady State and Transient FG Behavior
- Transient Fracture Behavior



### Engineering Scale Input

- Steady State Conditions
- Transient Conditions
- BISON Radial Distribution
  - Temperature
  - Burnup/Fission Rate

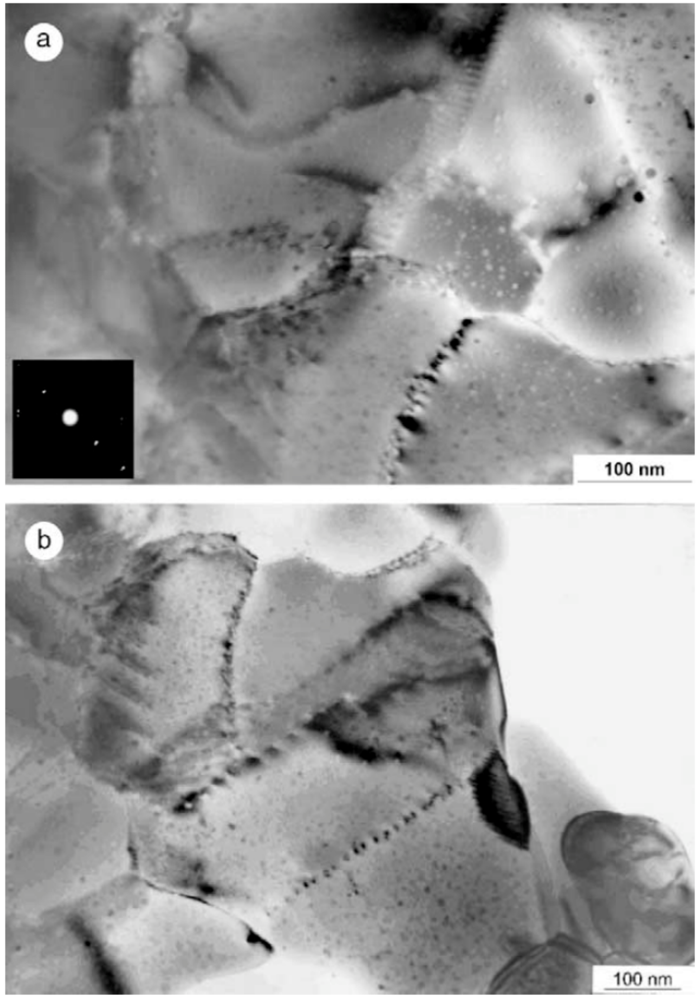
# Atomic Scale Pulverization Model Developed

*David Andersson* and *Michael Cooper*
*Los Alamos National Laboratory*

# Atomic scale simulation of fragmentation mechanisms

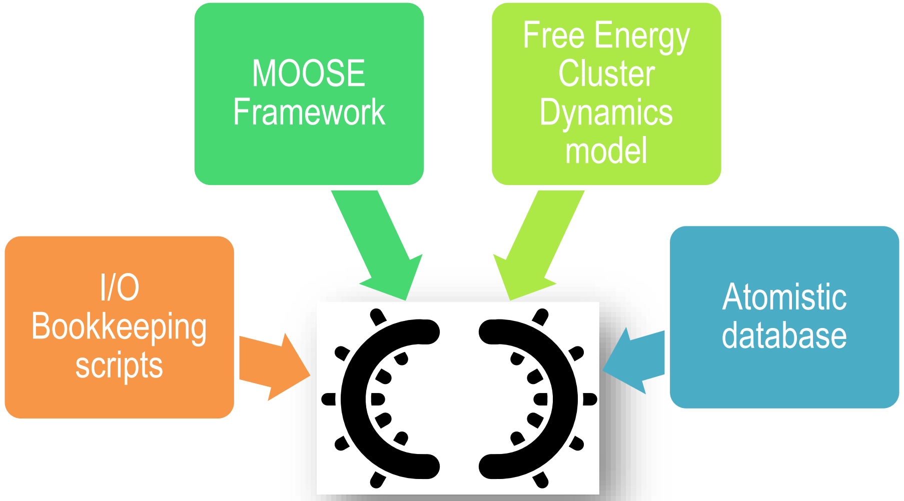
- HBS formation is characterized by coarsening of bubbles to micron-sized and sub-grain formation
- Experimental observation of nm-size bubbles decorating sub-grains (Sonoda et al., see right)
- Over-pressurized bubbles have been suggested as cause of fragmentation/pulverization of HBS during temperature 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? EPRI-funded

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



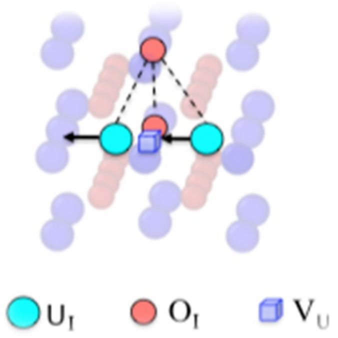

Sonoda et al. Nucl. Inst. Meth. Phys. Res. B 191 (2002) 622

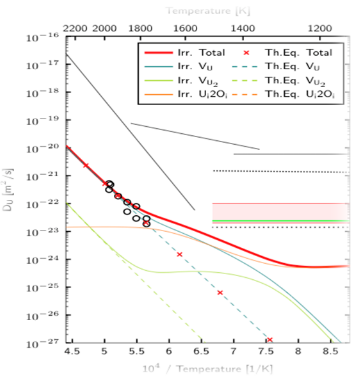
# Evolution of bubble pressure in HBS region




## "Cluster Dynamics Simulation of Uranium Self-diffusion During Irradiation in $UO_2$"

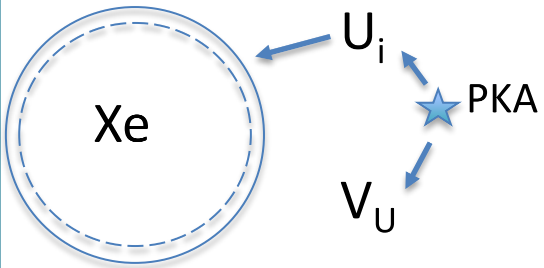
Matthews et al. JNM 527 (2019)




Used CD to calculate ***uranium defect*** cluster conc. and diffusivities under irradiation

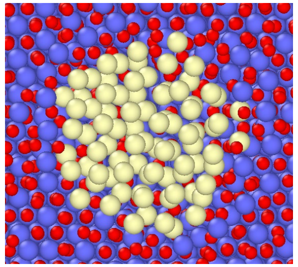
## Considering impact of defects on bubble pressure

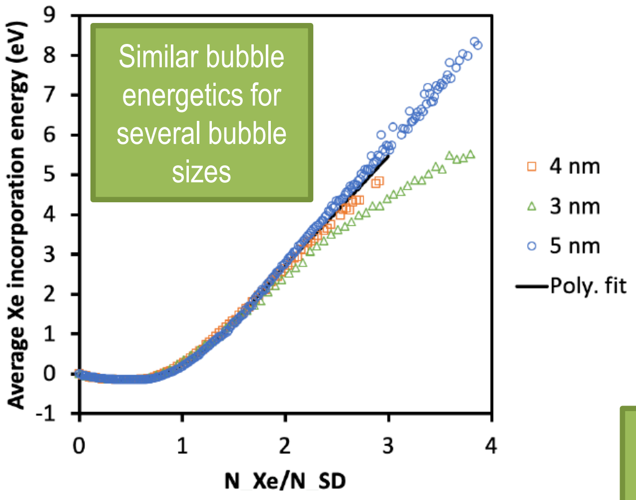



- When defects react with a bubble they change the pressure of the bubble
- The work done to change the bubble pressure needs to be included in the bubble-defect reaction energies
- We have used MD simulations to derive analytical expression of these new reaction energies

## MD-derived atomic scale data to capture bubble pressure effects

Xe bubbles are made of removed $UO_2$ units (SD) and incorporated Xe atoms. High pressure bubbles have a high Xe/SD ratio






$$\frac{E_{bubble} - E_{void}}{Xe} = f(Xe/SD)$$

$$\Delta E_{SD} = \frac{d(E_{bubble} - E_{void})}{dN_{SD}}$$

$$\Delta E_{Xe} = \frac{d(E_{bubble} - E_{void})}{dN_{Xe}}$$

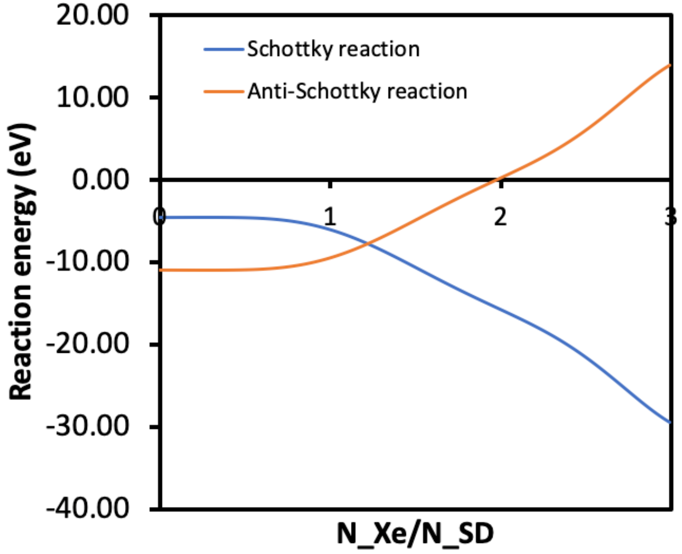
All as a function of Xe/SD only
Independent of bubble size

# Steady-state bubble pressure in HBS region

## New defect-bubble reaction energies

$$\{V_U : 2V_O\} \xrightarrow{G^{bubble}_{\{V_U:2V_O\}}} U_U + 2O_O + SD$$

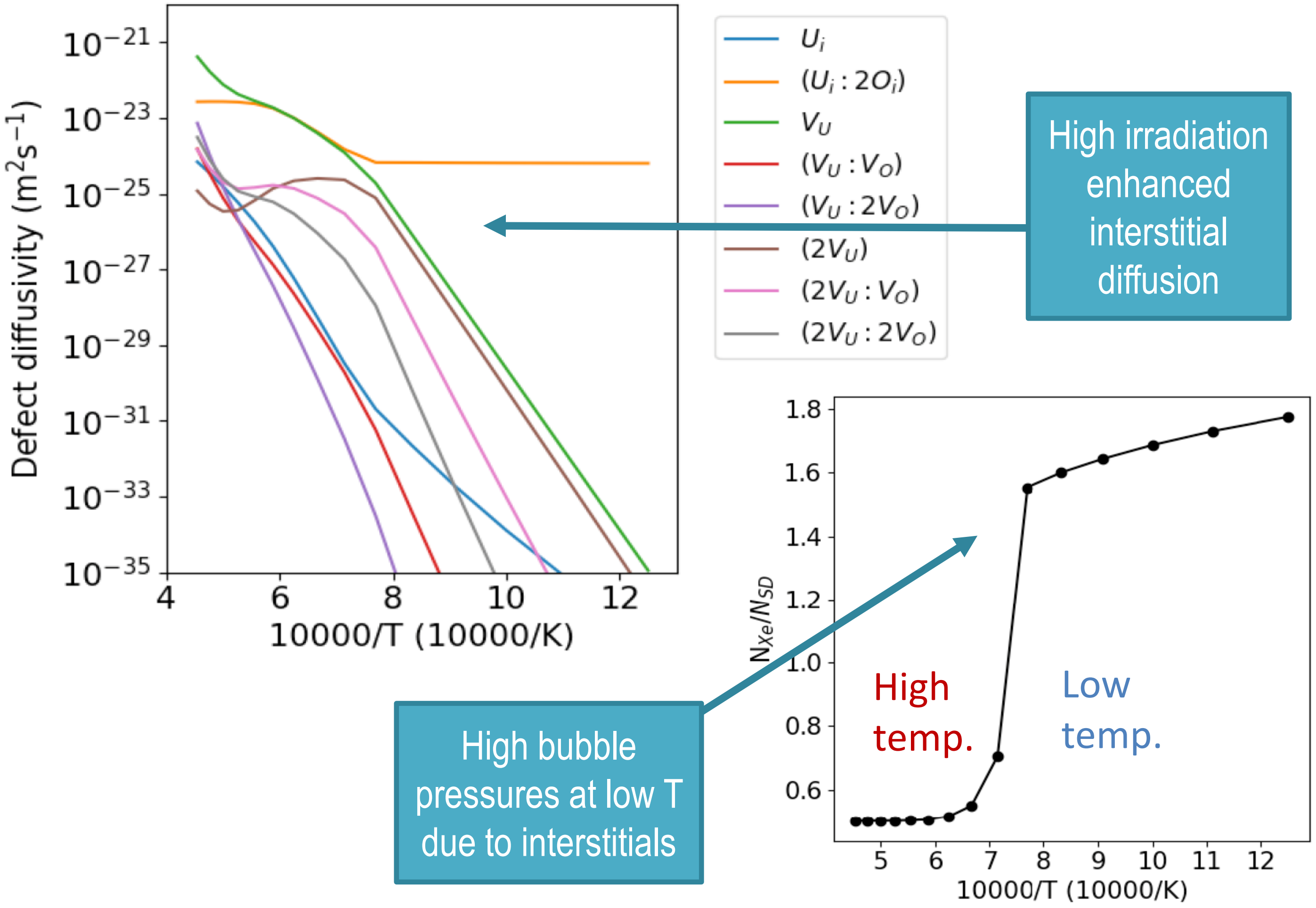
$$\{U_i : 2O_i\} + SD \xrightarrow{G^{bubble}_{\{U_i:2O_i\}}} 3V_i$$




Application of $\Delta E_{SD}$ 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*

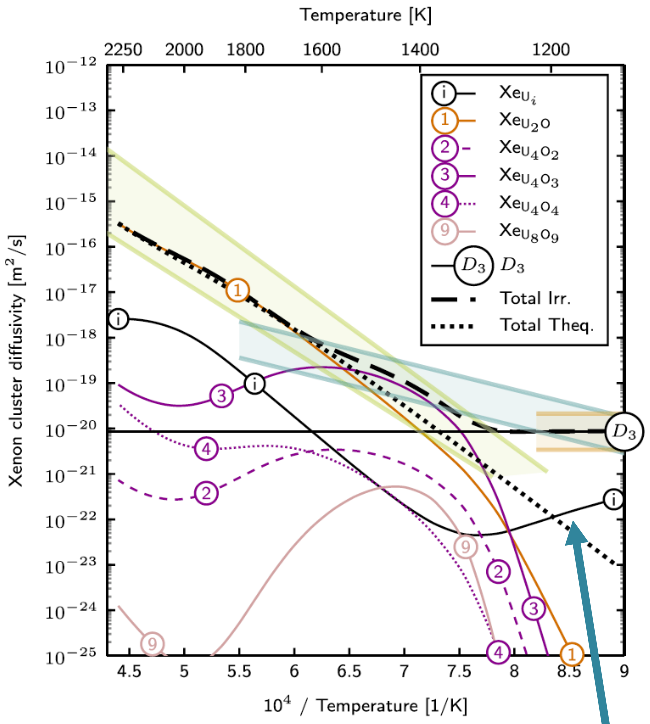
## Pressure-dependent Centipede simulations




*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

## Pressure-dependent Centipede simulations



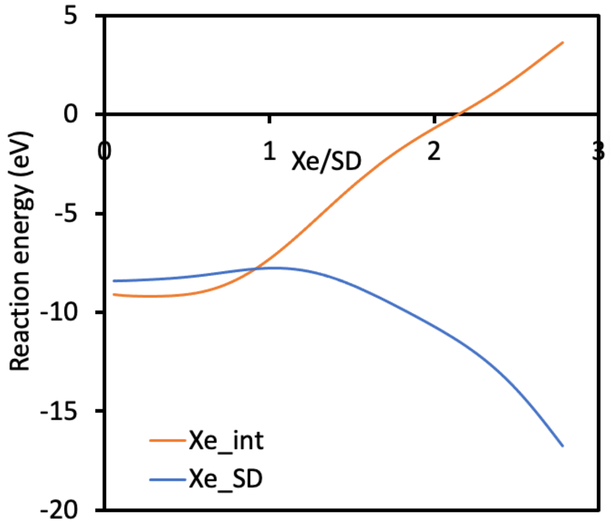

As for U diffusion Xe interstitials dominate at low T

"Cluster Dynamics Simulation of Uranium Self-diffusion During Irradiation in $UO_2$" Matthews at al. JNM 540 (2020) 152326

## New defect-bubble reaction energies

$$\{Xe_U : 2V_O\} \xrightarrow{G^{bubble}_{\{Xe_U:2V_O\}}} U_U + 2O_O + SD + Xe_{bubble}$$

$$Xe_i \xrightarrow{G^{bubble}_{Xe_i}} V_i + Xe_{bubble}$$

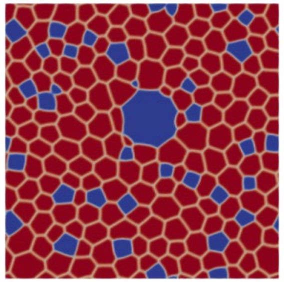



Application of $\Delta E_{SD}$ and $\Delta E_{Xe}$ to the reaction energies for Xe interstitials and Xe at vacancies with bubbles

*Next steps are to include Xe-sink reaction energies in Centipede*

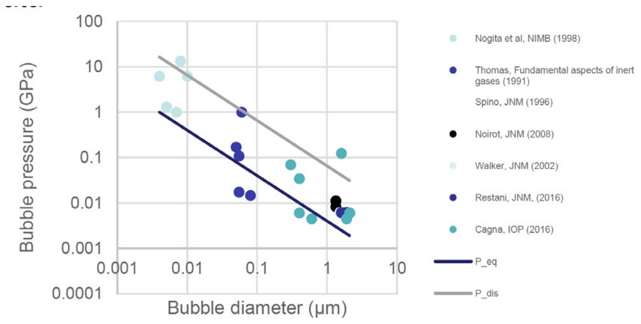
## Connection longer length scales and experiment

**Irradiation-enhanced interstitials expected to also be important at meso-scale. Collaborating on their implementation in phase field simulations**



(L Aagesen)

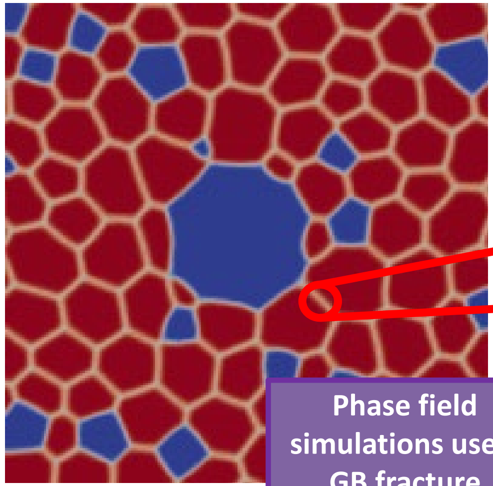
Potential development/use of new techniques to measure bubble pressure in HBS region under AFC will allow validation of our results




(F Cappia)

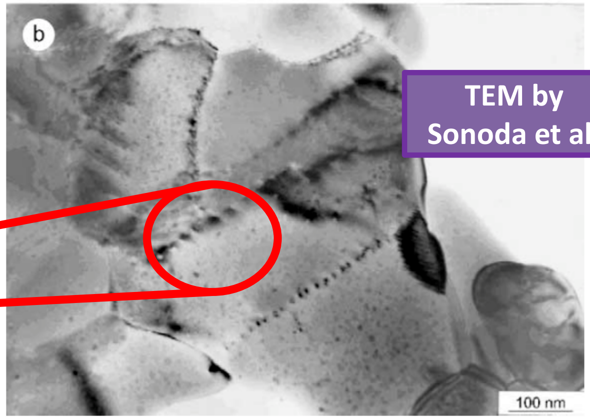
# Impact of high pressure bubbles on grain boundary strength

## Nm-size bubbles at sub-grains



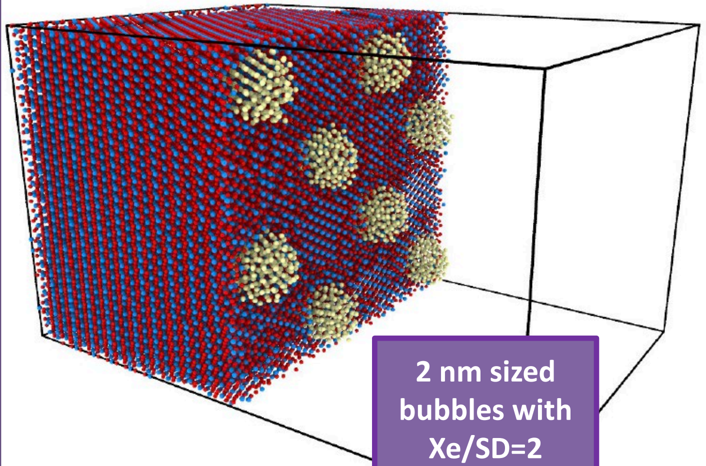
(L Aagesen)

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

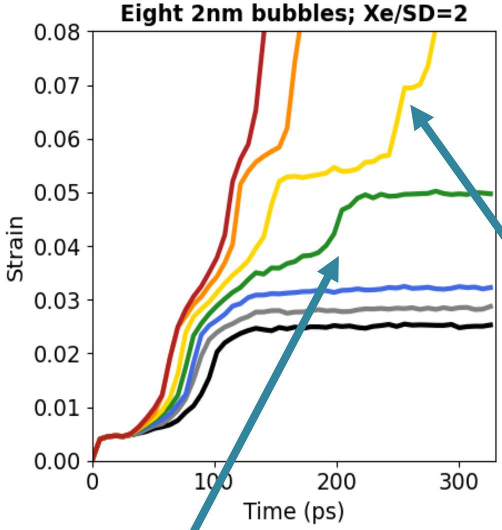


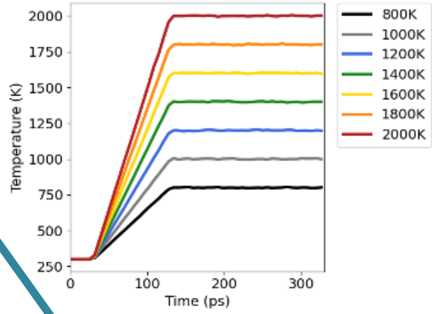
2 nm sized bubbles with Xe/SD=2

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.

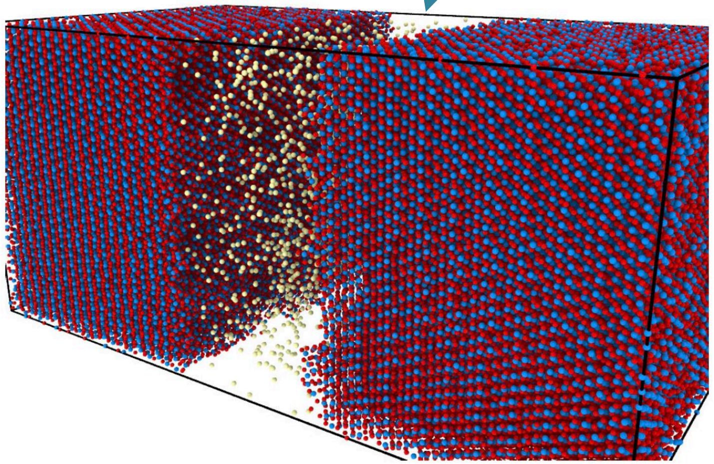
## Temperature ramp with no applied stress



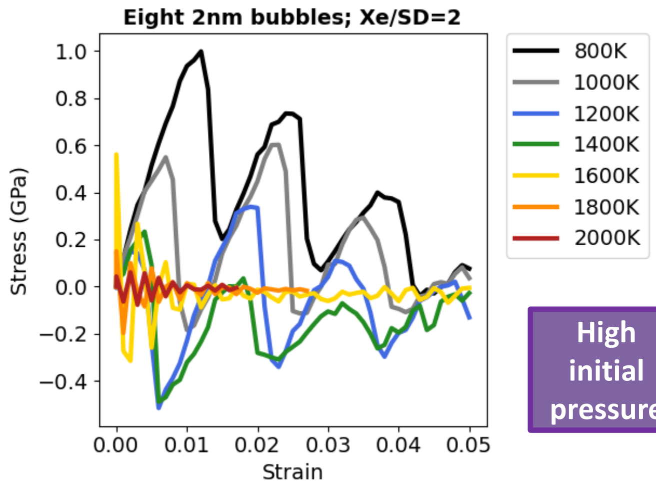

Evidence of micro-cracking

Spontaneous GB failure at high T

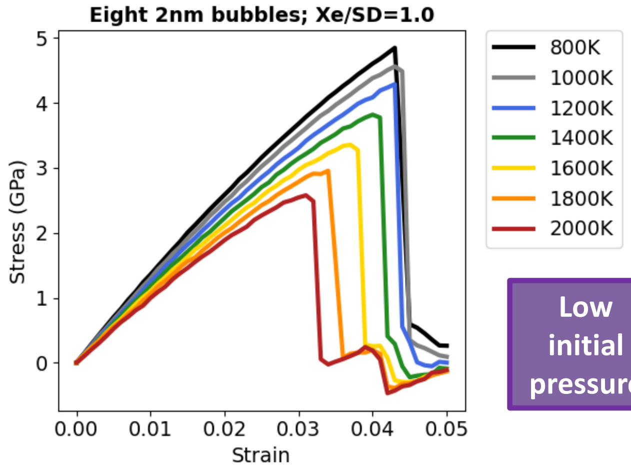


# Impact of high pressure bubbles on grain boundary strength

## Stress-strain analysis (impact of pressure)




High initial pressure




Low initial pressure

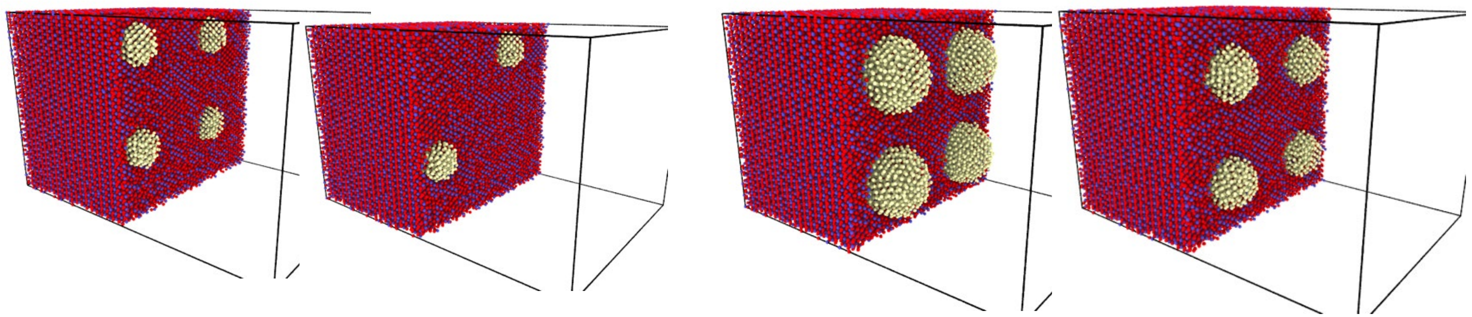
*High initial bubble pressures contributes to significant grain boundary weakening.*

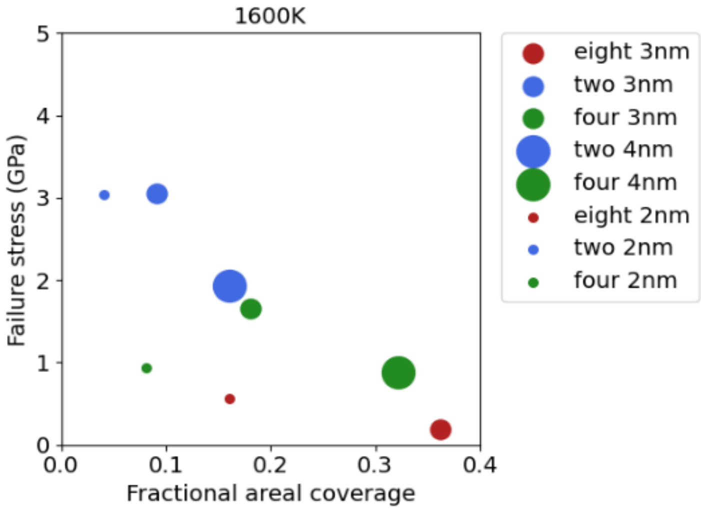
*Next steps under NEAMS are to further investigate intermediate pressures (1.5-2.0).*

## Stress-strain analysis (impact of areal coverage)

Varying number density

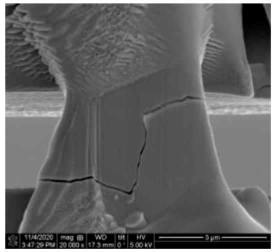
Varying size






The failure stress of the grain boundaries has been correlated to the bubble pressure, morphology, and the peak temperature.

**Failure criteria based on peak stress or energy release rate during fracture can be implemented in phase-field**



(F Cappia)

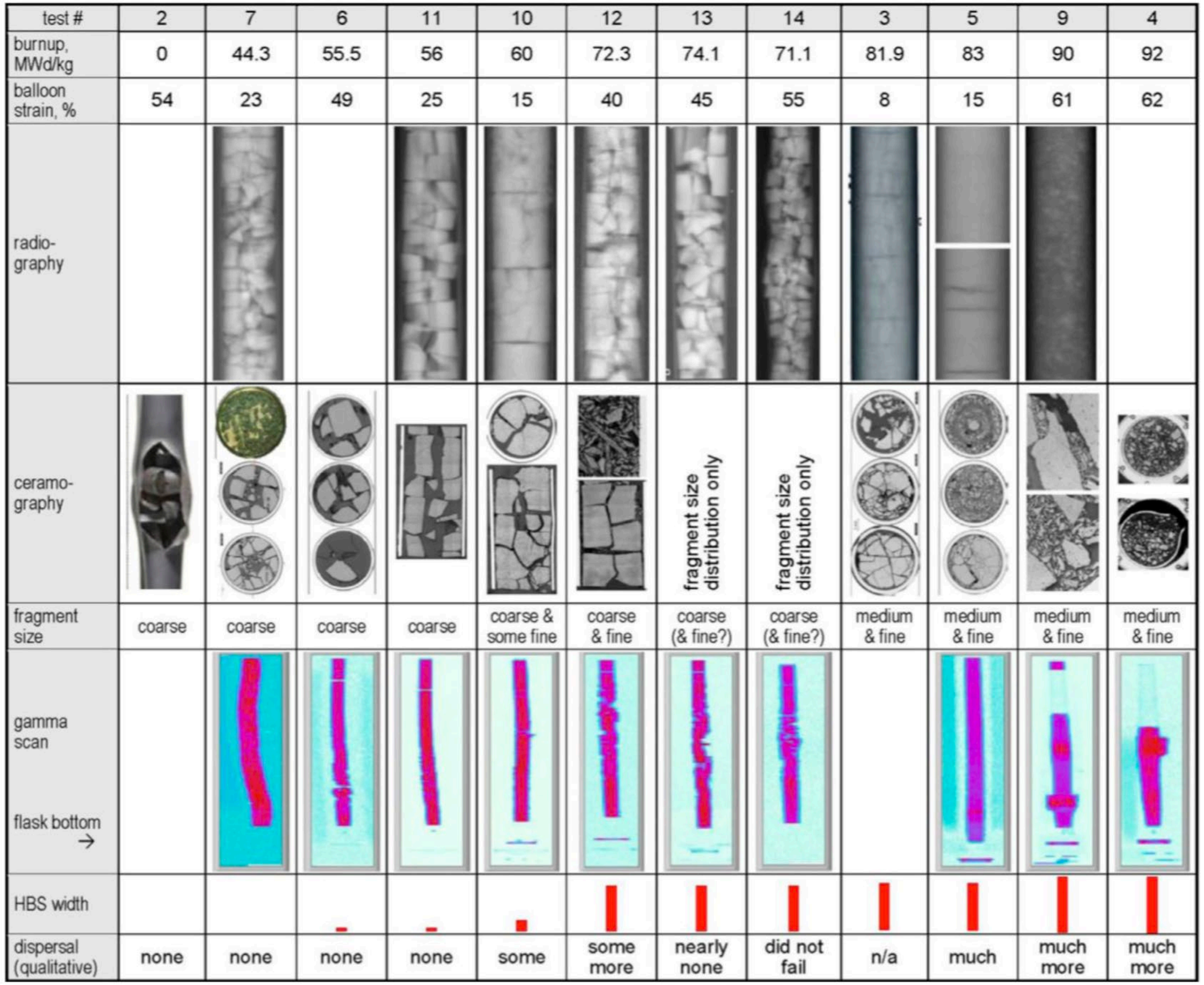
Results will be compared to ongoing PIE micro-mechanical tests under AFC.

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

# Material Model Needs to Address FFRD

- Phenomenon observed during Integral LOCA test
  - Rod Bending, Cladding Ballooning, Cladding Burst, Transient FGR, Rod Depressurization, Fuel Fragmentation and Pulverization, Fuel Relocation, and Dispersal

- BISON Model Development Needs
  - High Burnup $UO_2$ Pulverization
    - Transient FGR
    - High burnup structure
    - FG bubble nucleation, density, and pressure
  - High Temperature Cladding Creep
    - Anisotropic bending, ballooning, burst timing, burst opening, etc.
    - Application to LOCA and AOO events (i.e., margin recovery)



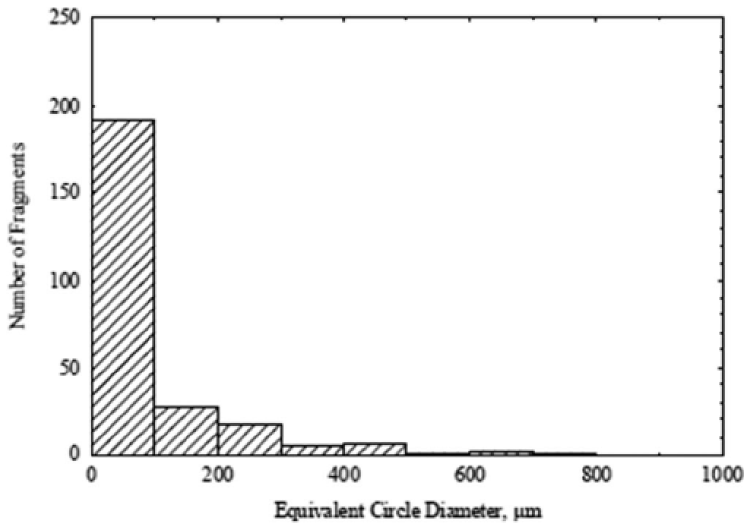
| test # | 2 | 7 | 6 | 11 | 10 | 12 | 13 | 14 | 3 | 5 | 9 | 4 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| burnup, MWd/kg | 0 | 44.3 | 55.5 | 56 | 60 | 72.3 | 74.1 | 71.1 | 81.9 | 83 | 90 | 92 |
| balloon strain, % | 54 | 23 | 49 | 25 | 15 | 40 | 45 | 55 | 8 | 15 | 61 | 62 |
| radio-graphy | | | | | | | | | | | | |
| ceramo-graphy | | | | | | | fragment size distribution only | fragment size distribution only | | | | |
| fragment size | coarse | coarse | coarse | coarse | coarse & some fine | coarse & fine | coarse (& fine?) | coarse (& fine?) | medium & fine | medium & fine | medium & fine | medium & fine |
| gamma scan<br>flask bottom → | | | | | | | | | | | | |
| HBS width | | | | | | | | | | | | |
| dispersal (qualitative) | none | none | none | none | some | some more | nearly none | did not fail | n/a | much | much more | much more |

# Mesoscale modeling of high burnup $UO_2$ fuel evolution and LOCA response

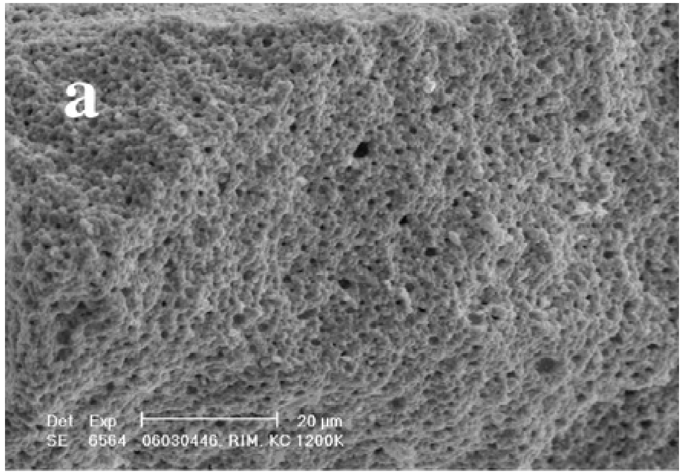
*Larry Aagesen*
*Idaho National Laboratory*

# Fine fragmentation/pulverization of high-burnup $UO_2$

- Potential to occur during loss-of-coolant accident (LOCA)-type temperature transients
- Formation of fine fragments <100 micron in size
- Fine particles can potentially escape into coolant from burst cladding during LOCA
- The phenomenon must be better understood to make a case for regulatory approval for higher burnup limits
- Hypothesized mechanism: During LOCA, trapped gas in bubbles heats up and becomes overpressurized; cracking initiates at these overpressurized bubbles



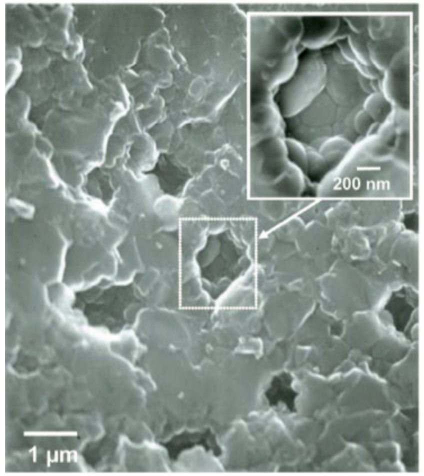

Turnbull et al., Nuc. Sci. & Eng., 179,477 (2015).




Hiernaut et al, JNM 377, 313 (2008).

# BISON model for pulverization

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

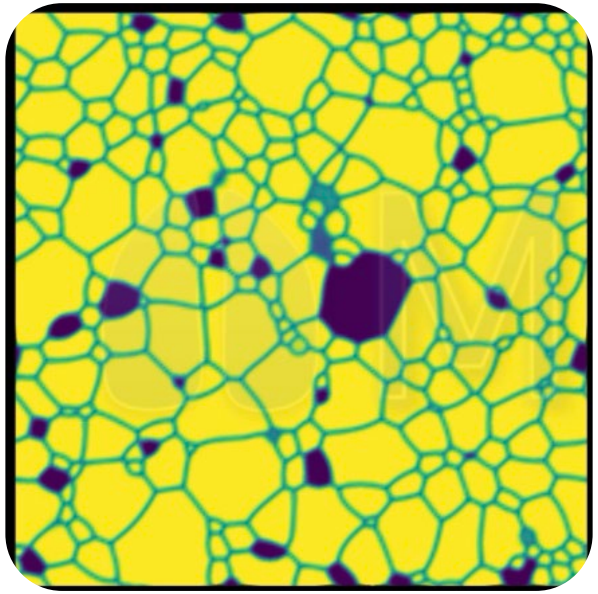



High-burnup structure in UO2
[Sonoda et al., NIMB, 2002].

- Current model is empirical, based on burnup and temperature
- Pulverization is predominantly observed to occur in regions where high-burnup structure (HBS) has partially or completely formed
  - HBS: Grain size decreases to ~150–200 nm, micron-sized bubbles form with multiple grains intersecting each bubble
- Goal: Develop a physics-based criterion for pulverization in BISON that accounts for microstructure
  - Focus on HBS
  - Mesoscale modeling of 3 stages: HBS formation, HBS bubble response to LOCA temperature transient, Fragmentation in fuel matrix surrounding bubbles

# Phase-field modeling of High Burnup Structure (HBS) formation

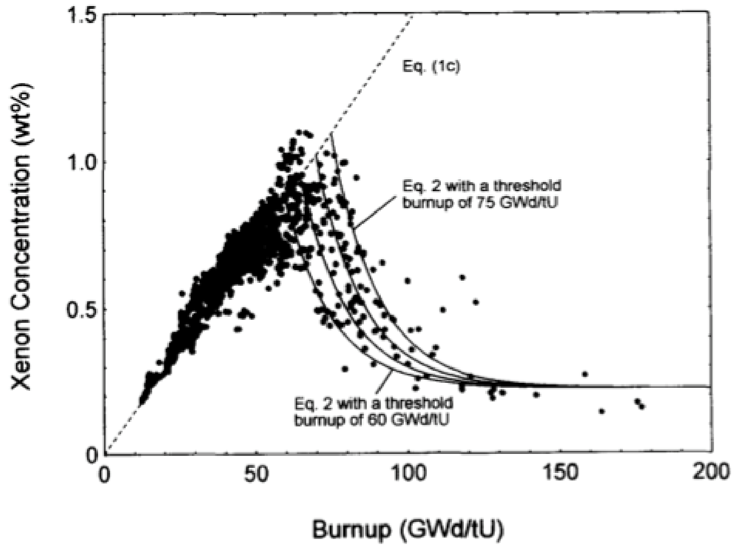
- Goals:
  - Understand mechanisms of HBS formation
  - Estimate initial gas pressures at beginning of LOCA
  - Provide microstructure as input for fracture simulations
- Multi-phase multi-order parameter model capable of capturing arbitrary number of phase, grains, and chemical components
- Nucleation model coupled with grand-potential based phase field model to demonstrate concurrent subgrain formation and bubble growth
- Defect species: vacancies and Xe atoms on vacancy sites
- Tracks dislocation density in each grain before and after recrystallization
  - Nucleation probability of recrystallized nuclei depends on local dislocation density and microstructural features



# Model Parameterization

- Defect source term based on for operating conditions. Diffusivities based on LANL atomistic calculations
- Vacancy sink is added to the model to match LANL-predicted steady-state vacancy concentration
- Initial super saturation value in the matrix is set based on the experimental data
- Pressure calculation has been added to the model

Van der Waals EOS: $$P_g = \frac{c_g\, k_B\, T}{\Omega - c_g b}$$




Lassman et al., JNM (1995)

**Xe Diffusion**

$$D_{tot}(T,\dot{F}) = D_1(T) + D_2(T,\dot{F}) + D_3(\dot{F}),$$

$$D_1(T) = \frac{2.22 \cdot 10^{-7} \exp(-3.26/k_B T)}{1 + 29.0 \exp(-1.84/k_B T)},$$

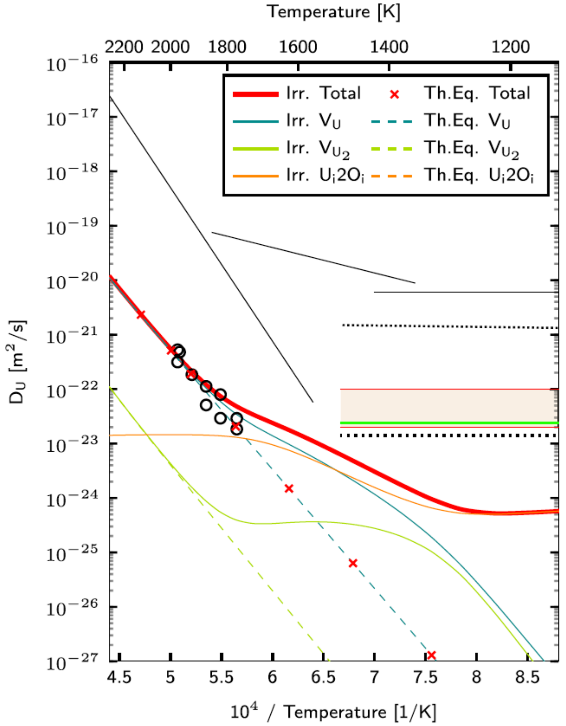
$$D_2(T,\dot{F}) = 2.82 \cdot 10^{-22} \exp(-2.0\,/\,k_B T)\sqrt{\dot{F}},$$

$$D_3(\dot{F}) = 8.5 \cdot 10^{-40} \dot{F},$$

Perriot et al, JNM (2019)

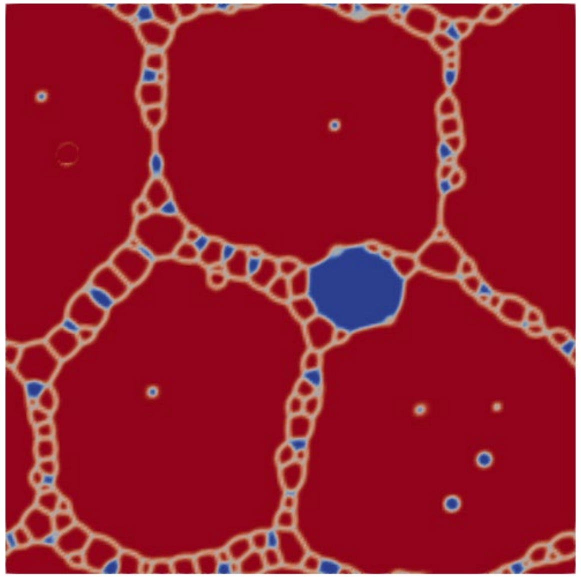
**U Diffusion**

Mathews et al., JNM (2020)



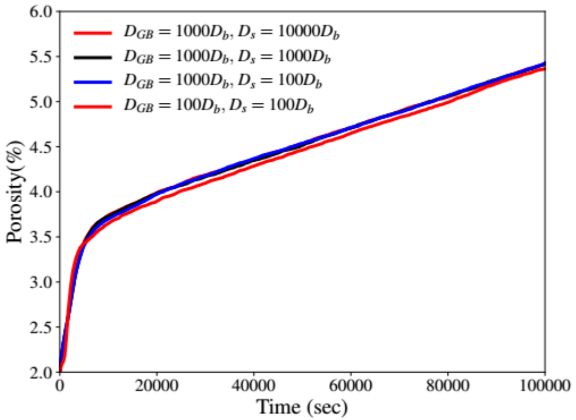
# Microstructure evolution during subgrain formation

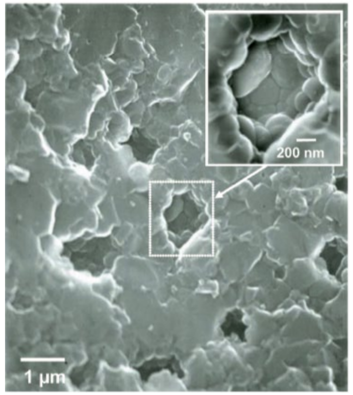
- Subgrain formation occurs due to dislocation density accumulation
- Defect accumulation causes formation of new bubbles or growth of existing bubbles
- Partial HBS formation is captured

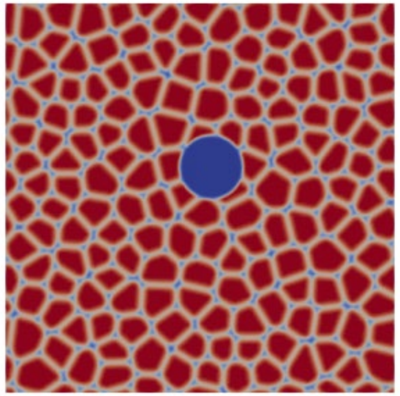


# Bubble evolution in HBS

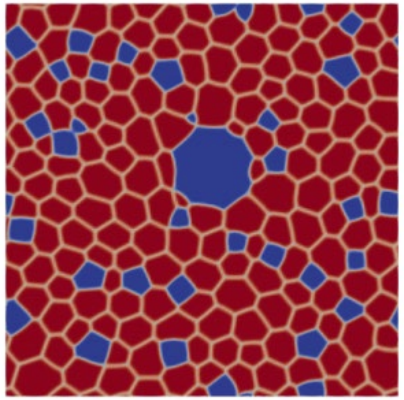
- Faster diffusion along GB and free surfaces facilitate formation and growth of large bubbles
- Bubble shape predicted from the simulation qualitatively matches the experimental observations



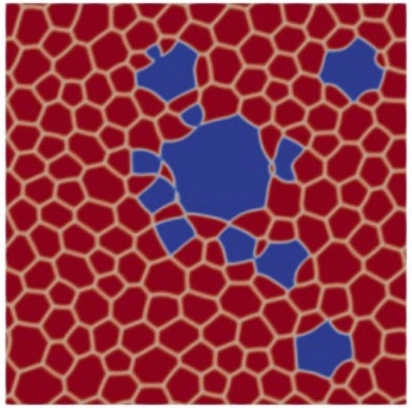



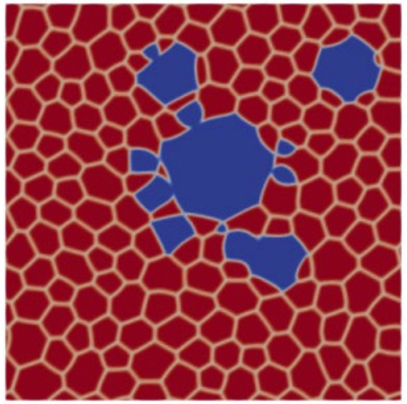
Initial Condition



DGB=0, Ds=0



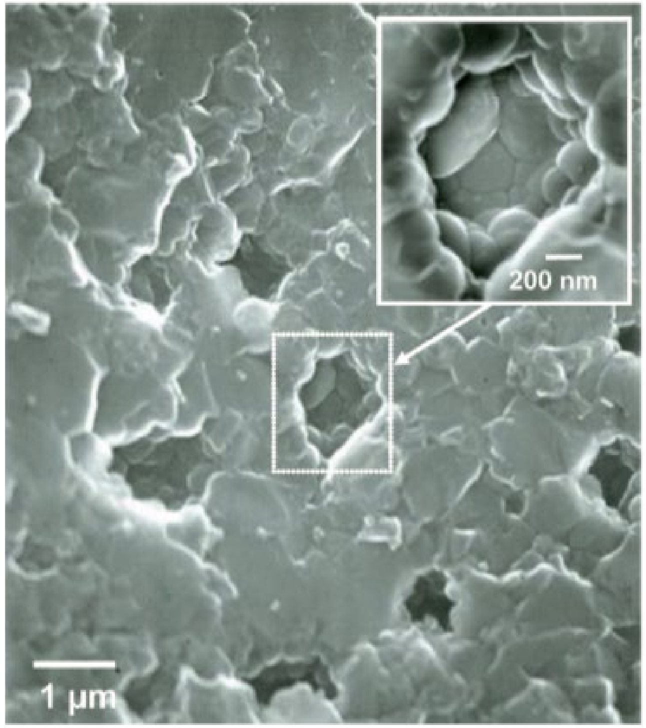
DGB=1000Dv, Ds= 100Dv



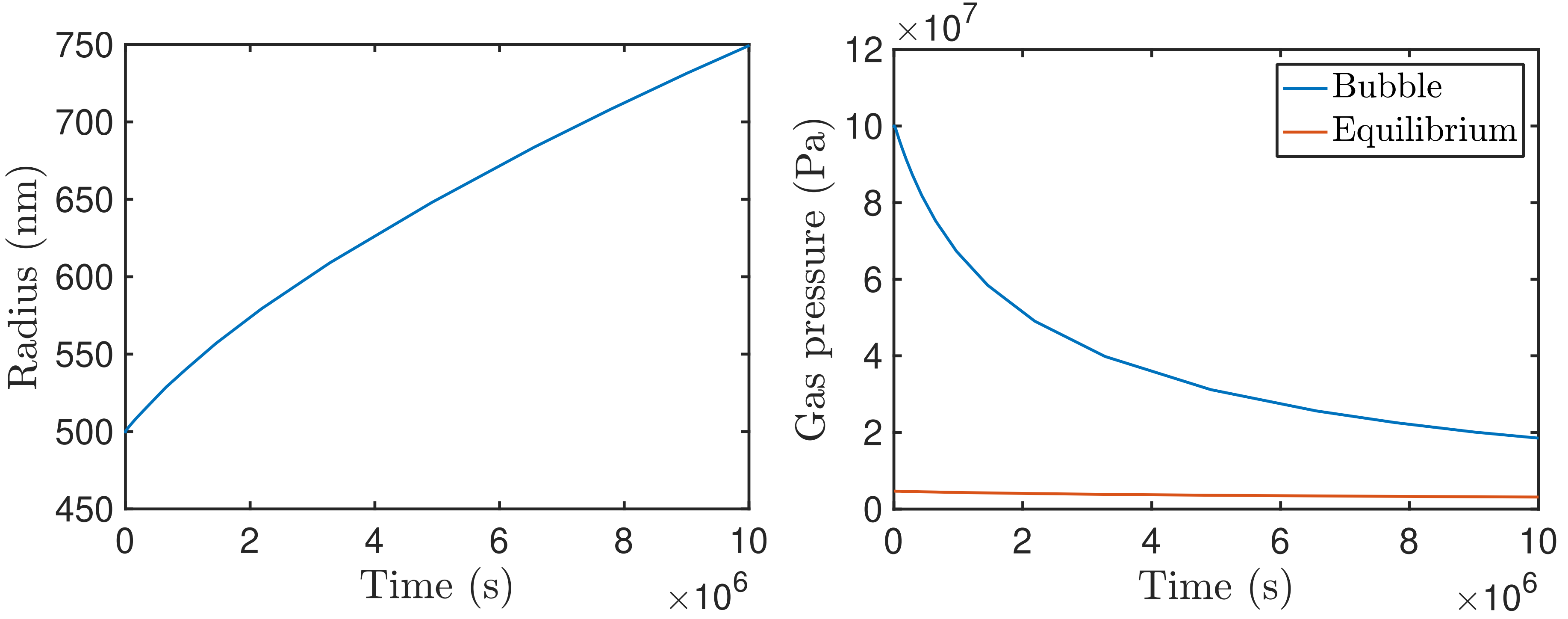
DGB=1000Dv, Ds= 1000Dv

# LOCA response of bubbles in High Burnup Structure (HBS) region

- Bubbles in HBS region: ~1 micron
- Believed to be significantly overpressurized relative to equilibrium given by Gibbs-Thomson equation
- Key Questions: During LOCA transient:
  - Does bubble size change significantly?
  - What does bubble pressure do? (inform PF Fracture)
- Development of new phase-field model to address LOCA behavior:
  - KKS formulation (removes bulk contribution to interfacial energy)
  - Use Helmholtz free energy, equation of state for van der Waals gas for bubble phase
  - Includes surface tension of bubble-matrix interface and gas pressure; allows consideration of effect of overpressurized bubbles



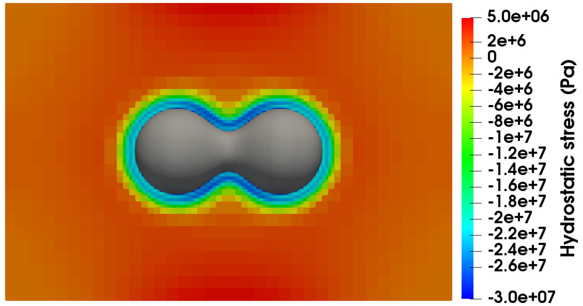
# Bubble growth during steady-state operation




- Parameterized with diffusivities and vacancy sink based on LANL calculations
- 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

# Bubble growth during steady-state operation

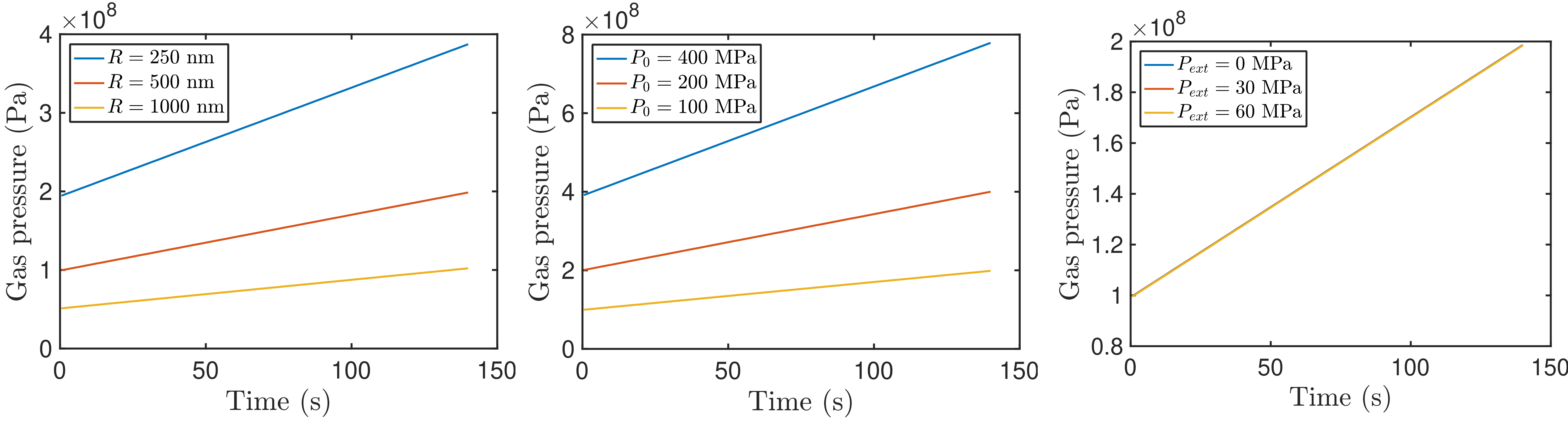
- 3D simulation to $1.5 \times 10^7$ s, 2 bubble impingement, initial radii of 300 nm
- Hydrostatic stress surrounding bubbles
  - Region of enhanced compressive hydrostatic stress in “neck”




$t = 1.3 \times 10^6$ s

# Bubble response to LOCA transient

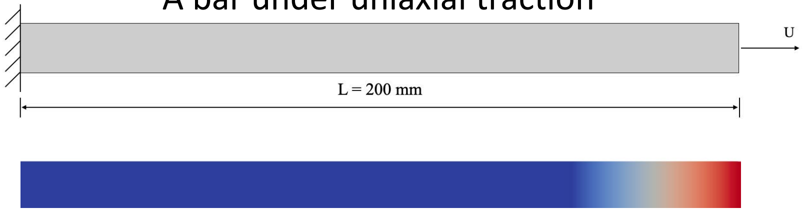
- Typical LOCA transient: start at 700 K, ramp 5K/s to final temperature 1400 K
- Consider variations in bubble size, initial pressure, porosity, external pressure
- **Bubble size does not change significantly for any cases considered**
- **Pressure as a function of time passed to PF fracture model**

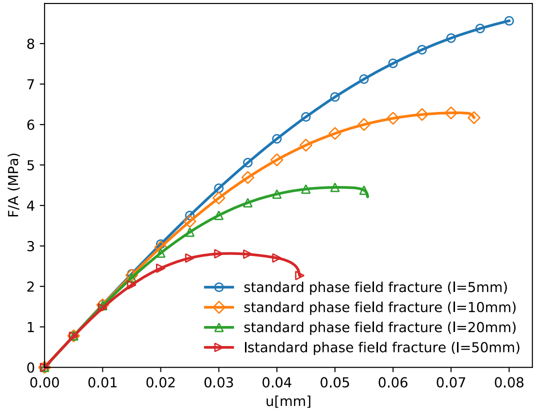


# Phase-field fracture simulation of HBS fuel

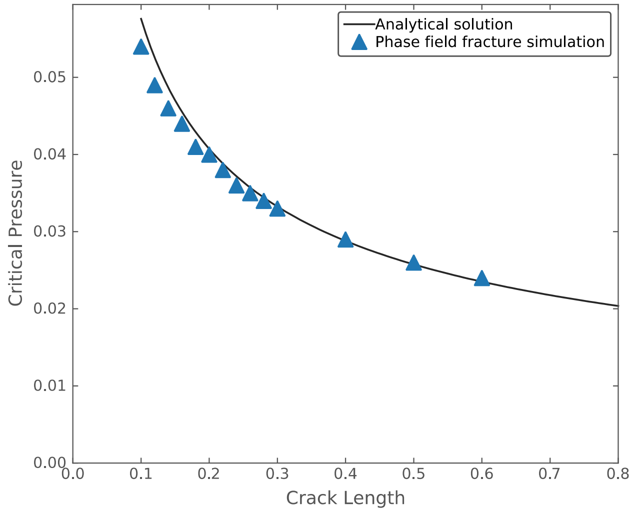
- Developed phase-field model for quasi-brittle fracture
  - It remains elastic behavior before crack initiates
  - Critical fracture strength is independent of length-scale parameter
  - It can predict general softening laws, such as linear softening law
- Developed phase-field model for pressurized 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

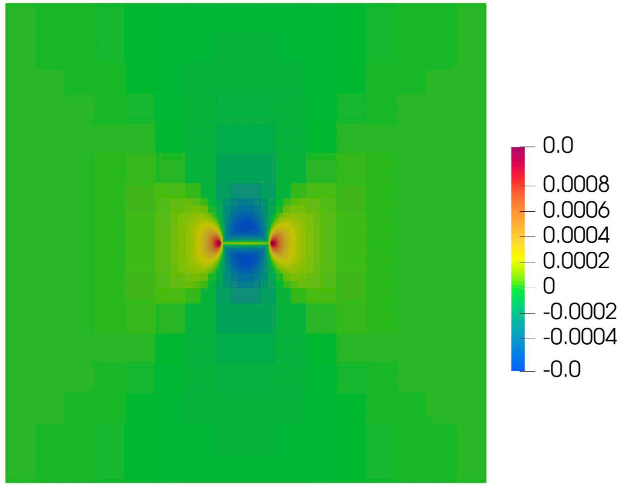


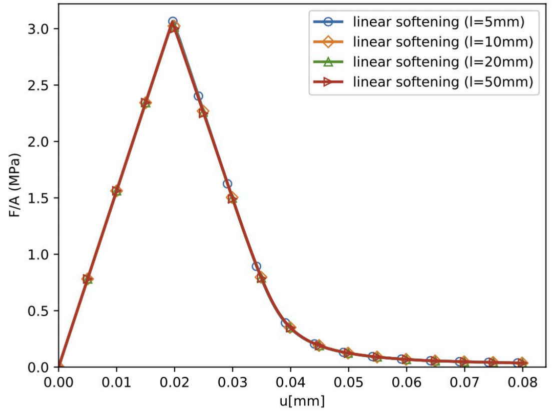

Classical brittle model




critical pressure vs. crack length




Stress contour of a pressurized crack

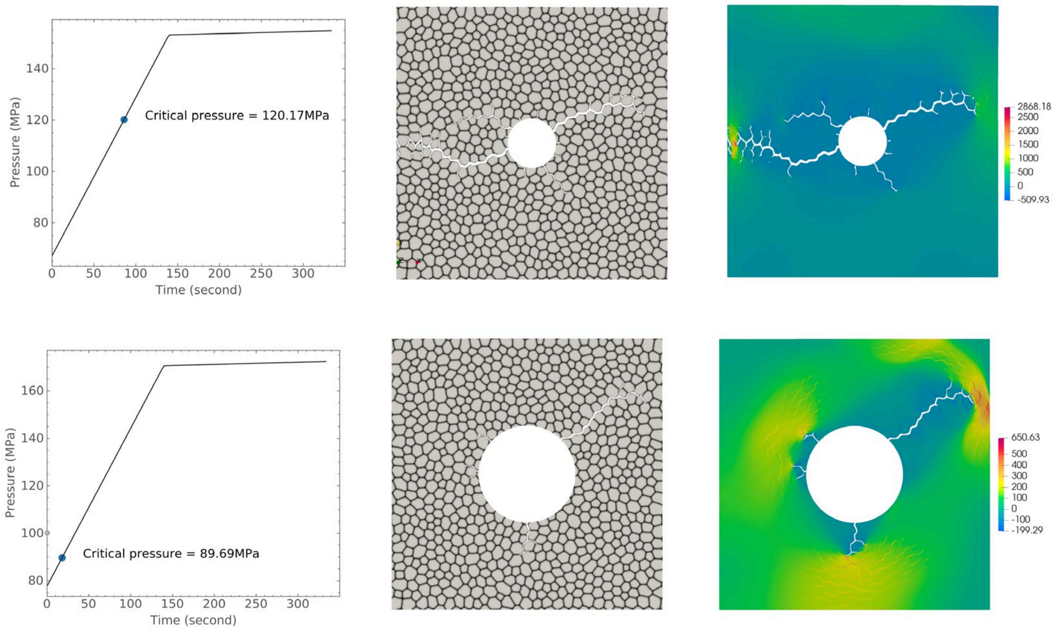



Quasi-brittle model

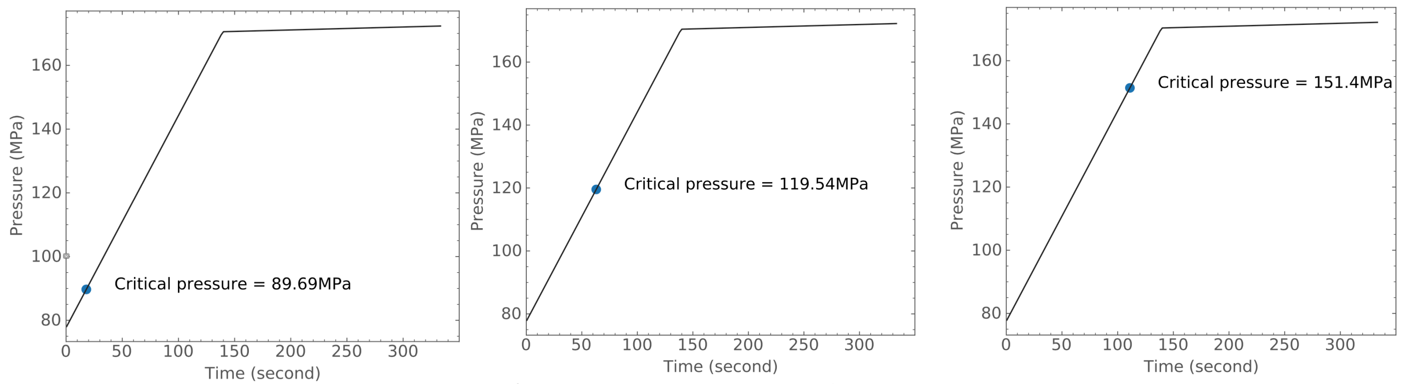
# Phase-field fragmentation modeling

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

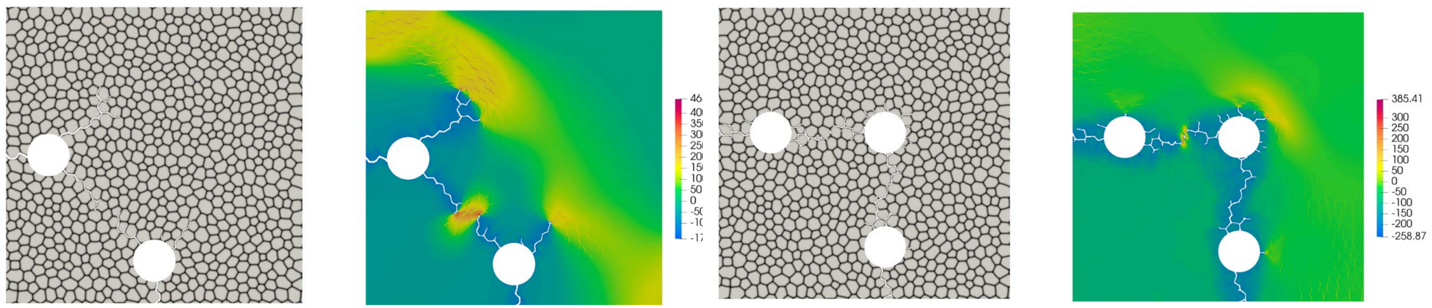
- Effect of bubble sizes: critical pressure is lower for the larger bubble
- Effect of external pressure: critical pressure becomes higher for larger external pressure values
- Effect of bubble interaction: fragmentation size is likely determined by bubble spatial distribution




Bubble size = 0.25 $\mu$m and 0.5 $\mu$m




External pressure = 0, 30 and 60 MPa

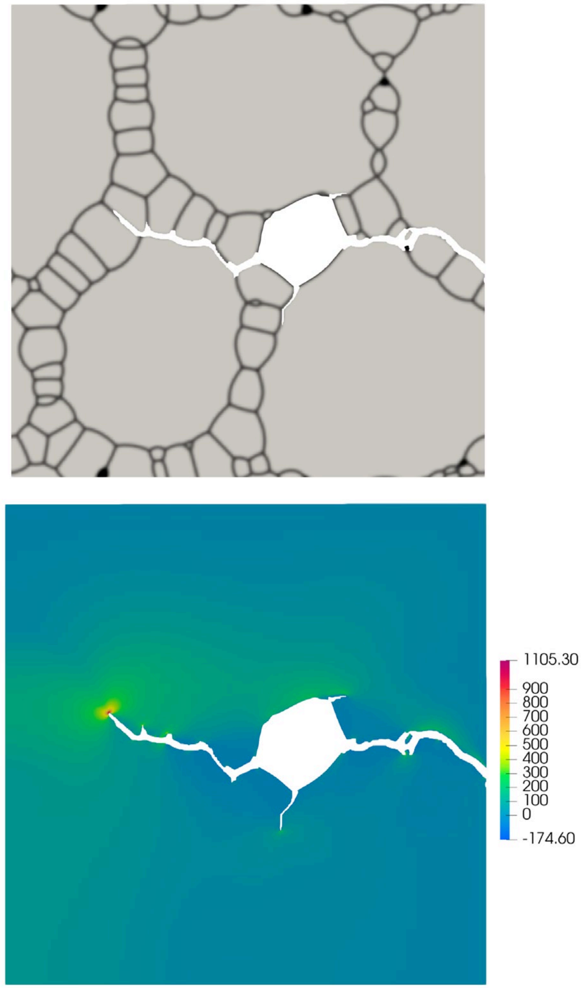


Two and three bubbles interaction

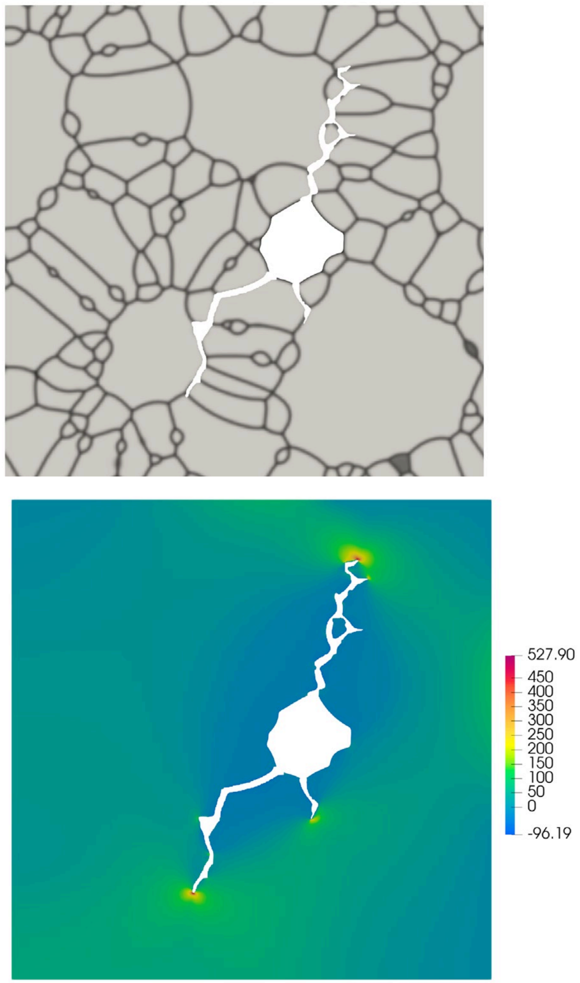
# Fragmentation in partial HBS

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

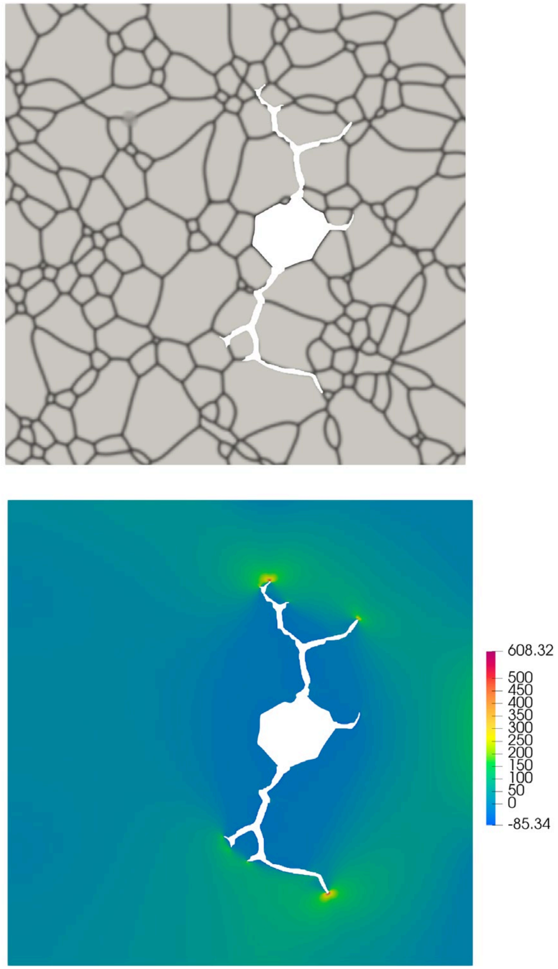
- Use the output from the HBS formation simulations as our initial condition
- Three HBS at different recrystallization stages with 25%, 60% and 100% recrystallization fraction were considered
- Crack initiation locations and crack propagation paths varied among the three cases because recrystallized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




25 % recrystallization stage



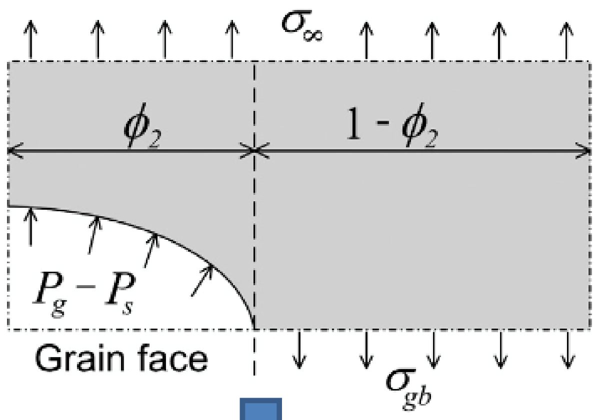

60 % recrystallization stage



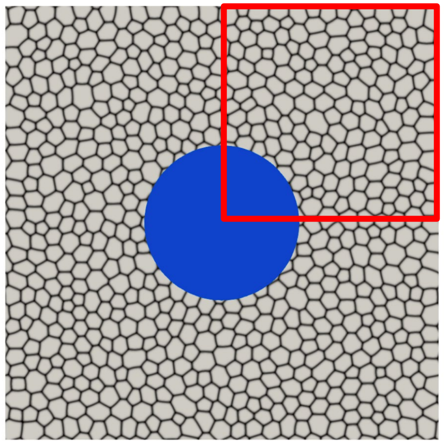

100 % recrystallization stage

# Physics-Based Criterion for Pulverization for BISON (Analytical)

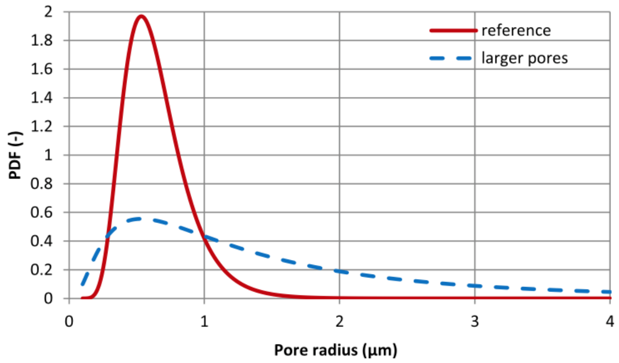
- Began implementation of pulverization based on analytical expression (Olander)
  - (To be supplanted by Phase-Field Fracture results)
- Originally developed for lenticular (non-HBS) bubbles
  - Adapted to HBS geometry using porosity, assume worst-case scenario of flat GB
- Pulverization occurs when $P_g > P_g^{cr}$
- Determine $P_g$ during transient for most frequently occurring bubble, $R$ = 0.53 $\mu$m
  - Initial pressure ~100 MPa at 673K based on experimental data
  - (To be supplanted by results from Phase-Field HBS formation model)




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



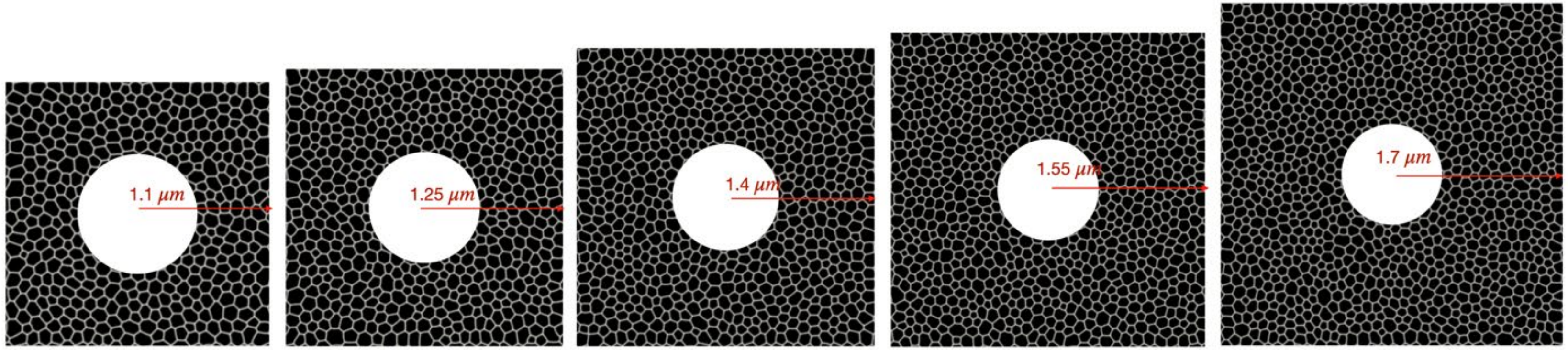
Determine $\phi_2$ from porosity (empirical)



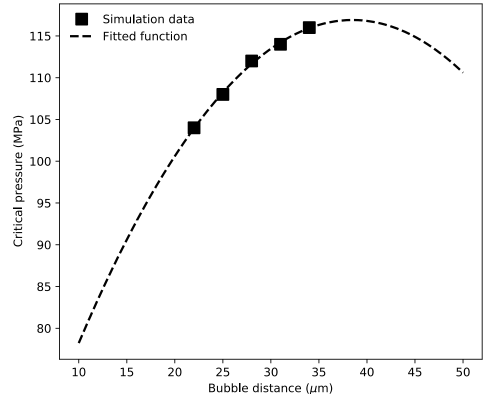

Kulacsy, JNM 466, 409-416 (2015)

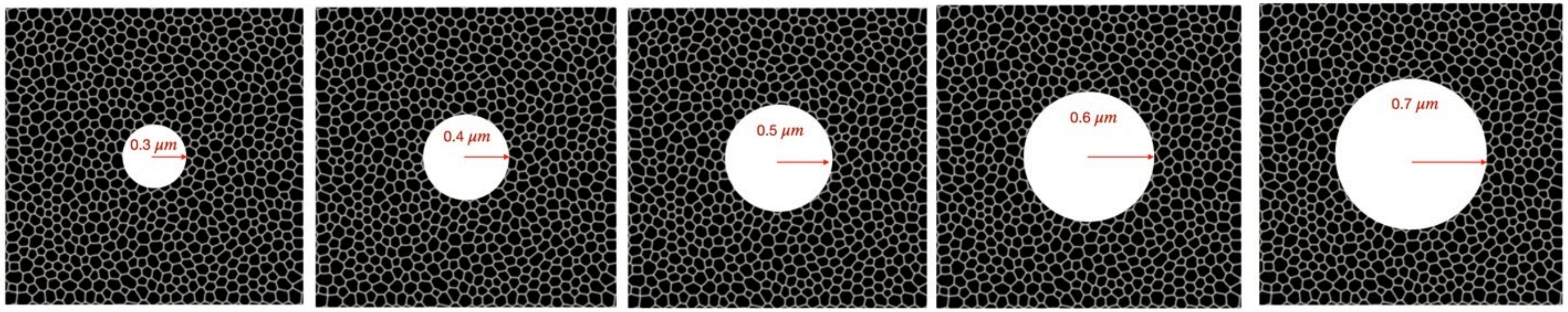
# Physics-Based Criterion for Pulverization for BISON (Simulation)

- Determine $P_g^{cr}$ from Phase-field fracture simulations with periodic boundary conditions to account for bubble-bubble interactions
- Pulverization occurs when $P_g > P_g^{cr}$
- Will also account for external pressure
- Determine $P_g$ during transient for most frequently occurring bubble

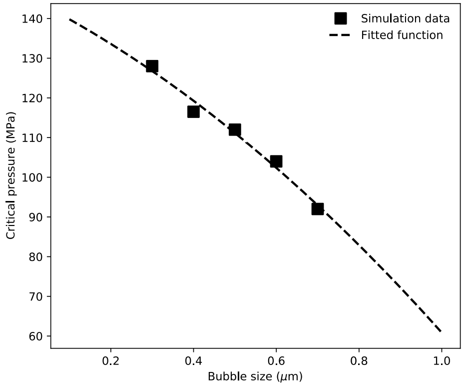



Initial configuration with different bubble distances




Initial configuration with different bubble sizes



# Data Needs to Support Material Model Development and Validation

- Integral LOCA and heat up test data
  - Cladding burst and balloon characteristics
  - Fuel Pulverization distribution
  - Porosity as a function of Radius
  - HBS transition as a function of Radius
  - Transient FGR
- Temperature at onset of pulverization

- Microstructure-level data
  - Gas bubble pressure (micron-size bubbles in HBS as well as nm-size bubbles at subgrain boundaries)
  - Fracture strength of subgrain boundaries in HBS (EPRI/AFC funded work at INL)

Questions?