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Phase-field simulations of fission gas bubbles in high burnup UO2 during steady-state and LOCA transient conditions

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Changing the World's Energy Future

Larry K Aagesen Jr, Sudipta Biswas, Wen Jiang, David Andersson, Michael Cooper, Christopher Matthews

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Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

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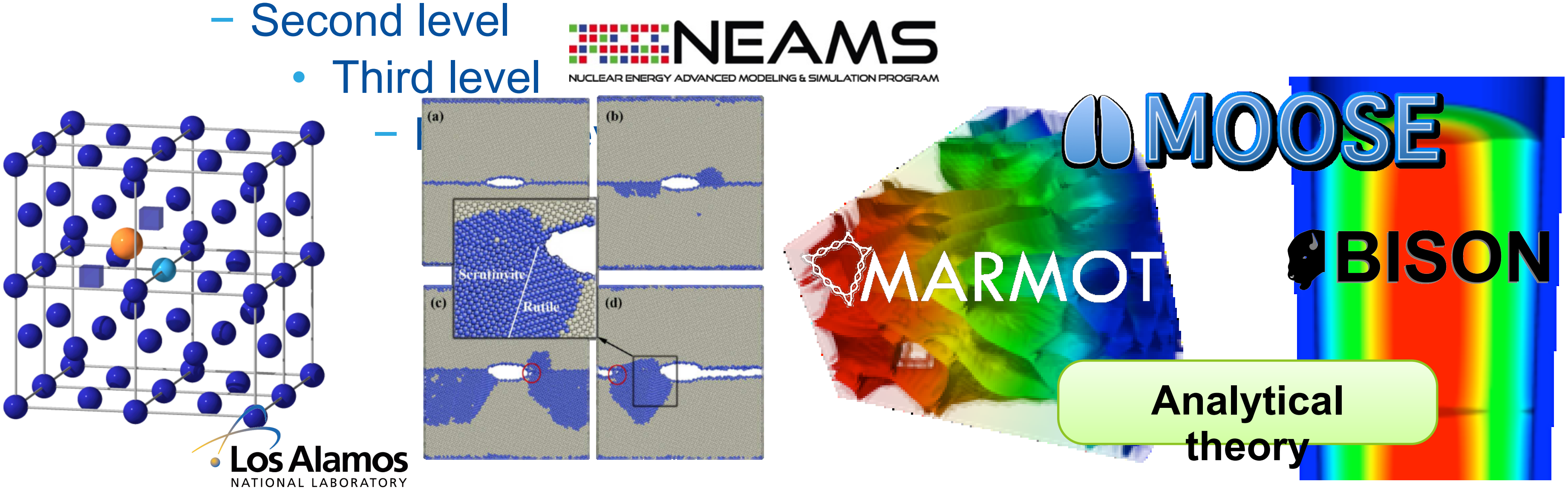
Phase-field simulations of fission gas bubbles in high burnup UO_2 during steady-state and LOCA transient conditions

Larry Aagesen
Idaho National Laboratory
Presented by David Andersson
Los Alamos National Laboratory
Wen Jiang, Sudipta Biswas
Idaho National Laboratory
Michael Cooper, Topher Matthews
Los Alamos National Laboratory

Idaho National Laboratory

Multiscale fuel performance simulation

- BISON: Fuel performance code developed at INL
- Inform BISON with atomistic and mesoscale simulations
  - Marmot: MOOSE-based phase-field simulation code



nanometers
First Principles
- Identify critical bulk mechanisms
- Determine bulk properties

100's of nanometers
Molecular Dynamics
- Identify interfacial mechanisms
- Determine interfacial properties

microns
Mesoscale
- Predict microstructure evolution
- Determine impact on properties

millimeters and up
Engineering Scale
- Use analytical theory
- Predict fuel performance

Fine fragmentation/pulverization of high-burnup UO_2

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- 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
- Industry would like to understand this problem better to strengthen their case for increasing fuel burnup limits
- Hypothesized mechanism: During LOCA, trapped gas in bubbles heats up and becomes overpressurized; cracking initiates at these overpressurized bubbles

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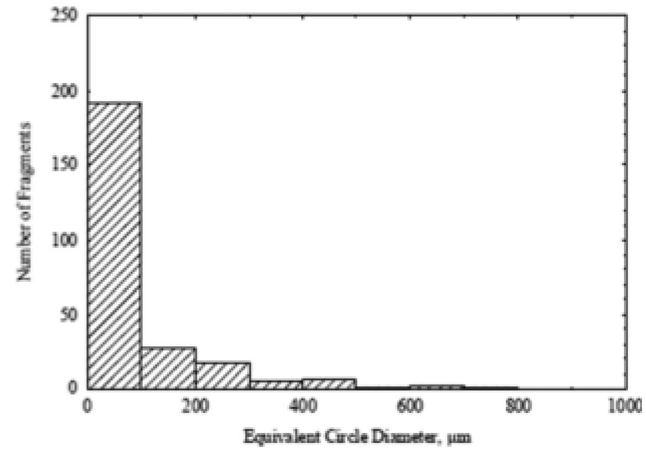
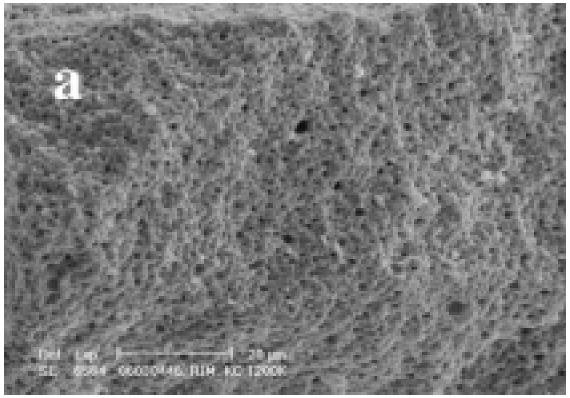


Turnbull et al., Nuc. Sci. & Eng., 179,477 (2015).



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

BISON model for pulverization

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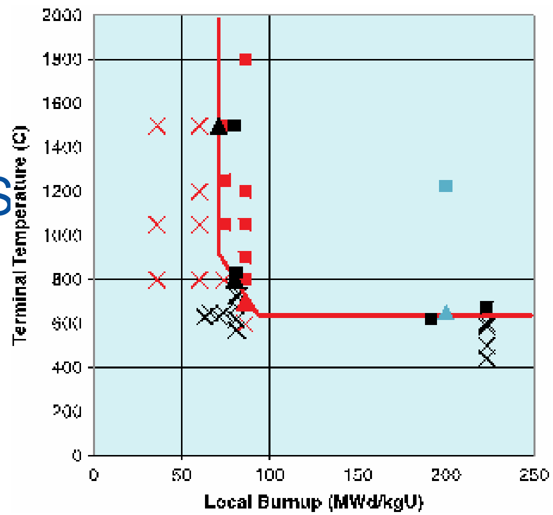

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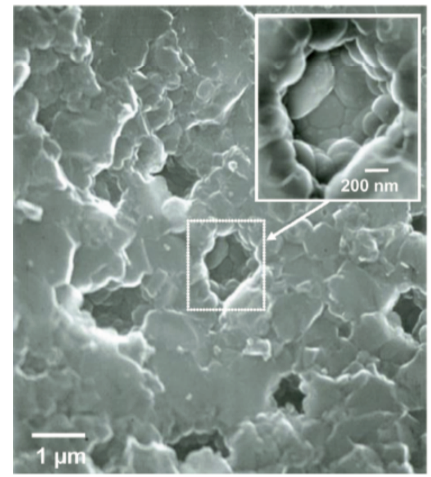
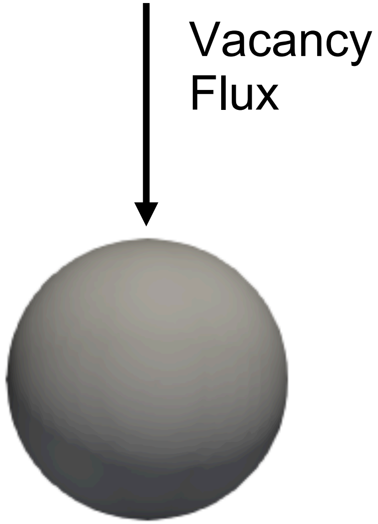


High-burnup structure in UO2 [Sonoda et al., NIMB, 2002].

- 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

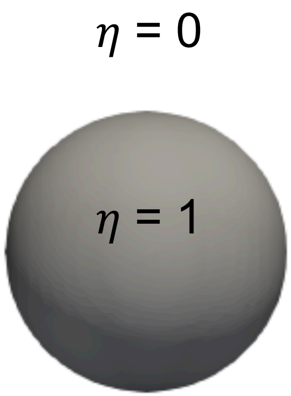
HBS bubble response to LOCA transient

- Bubbles in HBS region are ~1 μm and believed to be overpressurized relative to equilibrium (based on observed dislocation punching around bubbles):
- 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?**



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



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Phase-field model: Free energy functional

$$F = \int_V \left[f_{chem} + W g(\eta) + \frac{\kappa}{2} |\nabla \eta|^2 + f_{el} \right] dV$$

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- bulk chemical free energy density. $h(\eta)$ is a smooth interpolation function:

$$f_{chem} = [1 - h(\eta)] f^m_{chem}(c^m_g, c^m_v) + h(\eta) f^b_{chem}(c^b_g, c^b_v)$$

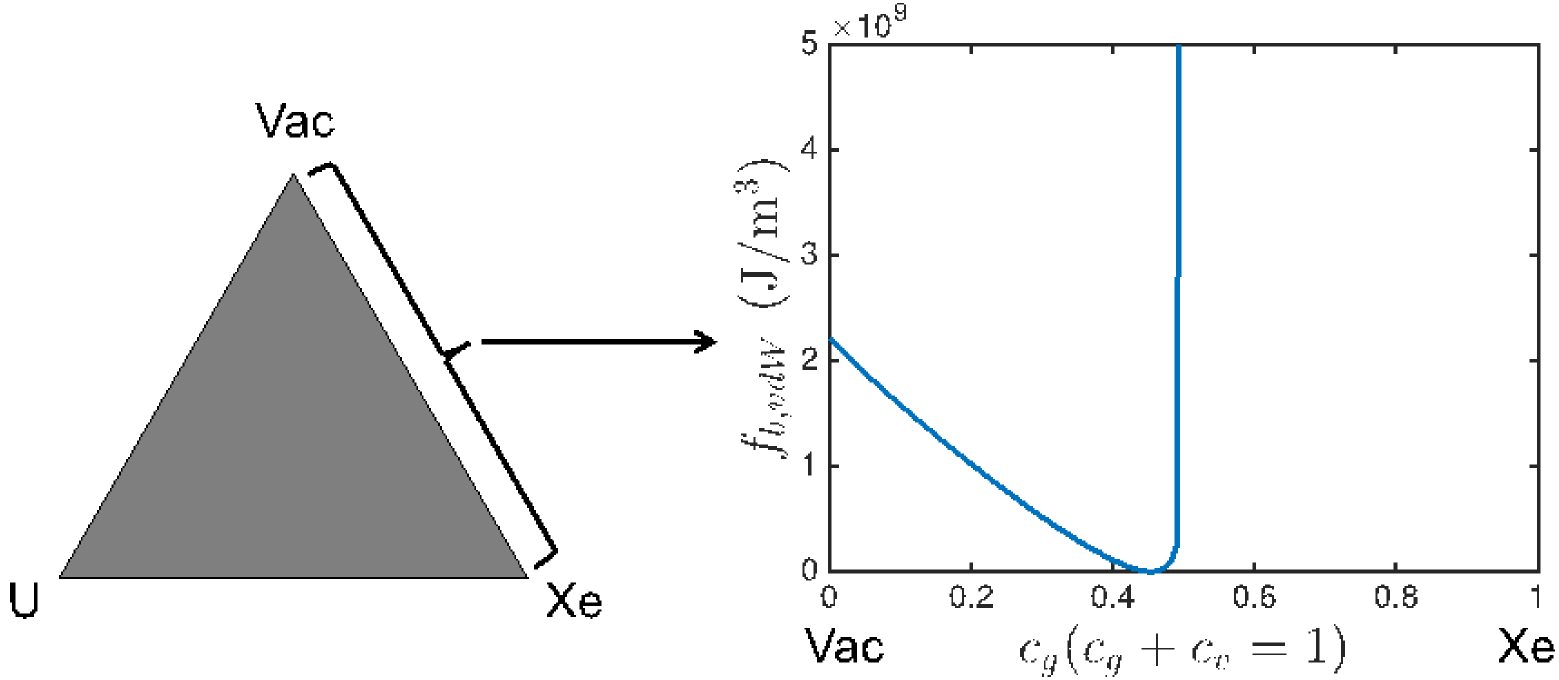
- chemical free energy of the matrix phase. Fit a parabolic approximation to ideal solution energetics:

$$f^m_{ideal} \approx f^m_{chem} = \frac{k^m_v}{2}(c^m_v - c^{m,eq}_v)^2 + \frac{k^m_g}{2}(c^m_g - c^{m,eq}_g)^2$$

- chemical free energy of the bubble phase. The bubble is considered to be a mixture of gas atoms and U-site vacancies. Energy given by the Helmholtz free energy of a van der Waals gas:

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

Parameterization for Xe gas phase



Gibbs triangle: U lattice sites

Helmholtz free energy density: Van der Waals gas

Phase-field Model: Elastic energy

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- Interpolating elastic energies and stresses (Voigt-Taylor scheme):

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$$f_{el} = [1 - h(\eta)] f_{el}^{m} + h(\eta) f_{el}^{b} \qquad f_{el}^{m} = \frac{1}{2} C_{ijkl}^{m} \epsilon_{ij}^{el,m} \epsilon_{kl}^{el,m} \qquad f_{el}^{b} = \frac{1}{2} C_{ijkl}^{b} \epsilon_{ij}^{el,b} \epsilon_{kl}^{el,b}$$

- Mechanical equilibrium equation:

$$\nabla \cdot \sigma_{ij} = \nabla \cdot [(1 - h(\eta)) \sigma_{ij}^{m} + h(\eta) \sigma_{ij}^{b} + \sigma_{ij}^{st}] = 0 \qquad \sigma_{ij}^{m} = C_{ijkl}^{m} \epsilon_{kl}^{el,m}$$

- Eigenstrain due to vacancies:

$$\epsilon_{ij}^{el,m} = \epsilon_{ij} - \epsilon_{ij}^{0} = \epsilon_{ij} - (c_v - c_v^{eq}) \epsilon_v^{0} \delta_{ij}$$

- Bubble pressure:

$$\sigma_{ij}^{b} = -\left(\frac{kT}{\frac{V_a}{c_g} - b} \right) \mathbf{I} + C_{ijkl}^{b} \epsilon_{kl}^{el,b}$$

- Surface tension:

$$\sigma_{ij}^{st} = \left[W g(\eta) + \frac{\kappa}{2} |\nabla \eta|^2 \right] \mathbf{I} - \kappa \nabla \eta \otimes \nabla \eta$$

Evolution equations

- Allen-Cahn for order parameter:

$$\frac{\partial \eta}{\partial t} = -L\left[\frac{\partial F}{\partial \eta}\right]$$

$$= -L\left[\frac{\partial h}{\partial \eta}(f_b - f_m) - \mu_v(c_v^m - c_v^b) - \mu_g(c_g^m - c_g^b)\right] - W\frac{dg}{d\eta} - \kappa\nabla^2\eta\Bigg]$$

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

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

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

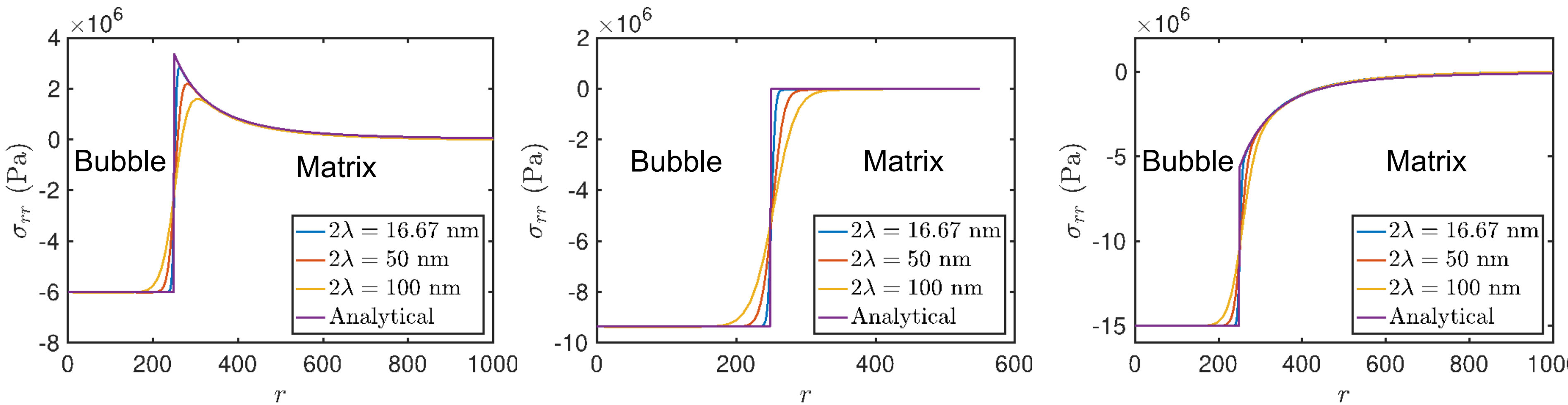
- Mobilities are a function of defect diffusivities:

$$M_v = \frac{h D_v^b + (1-h) D_v^m}{\partial^2 f / \partial c_v^2} \qquad M_g = \frac{h D_g^b + (1-h) D_g^m}{\partial^2 f / \partial c_g^2}$$

- + KKS system constraints

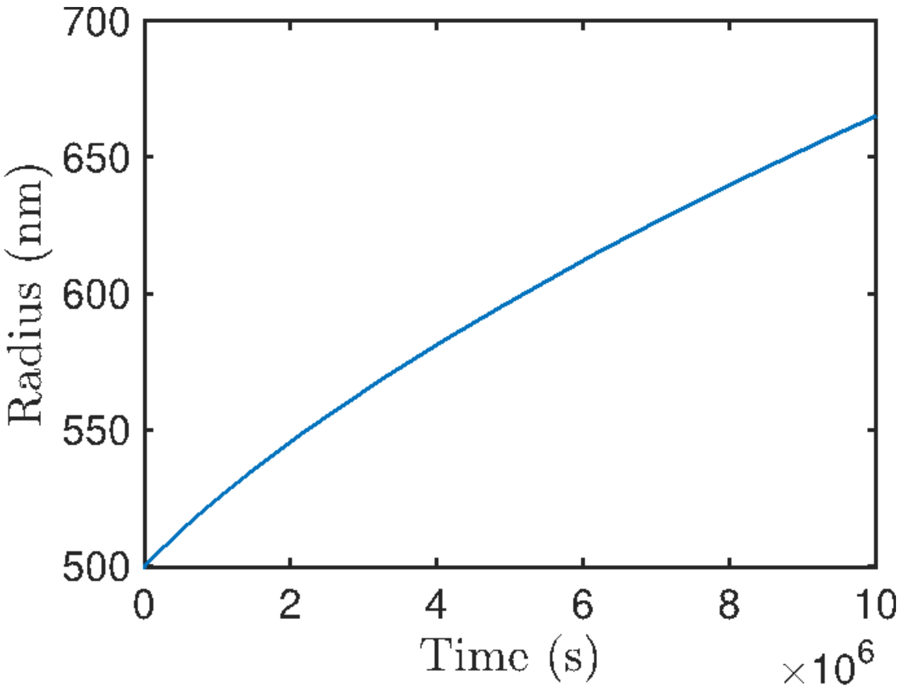
Phase-field Model verification and testing

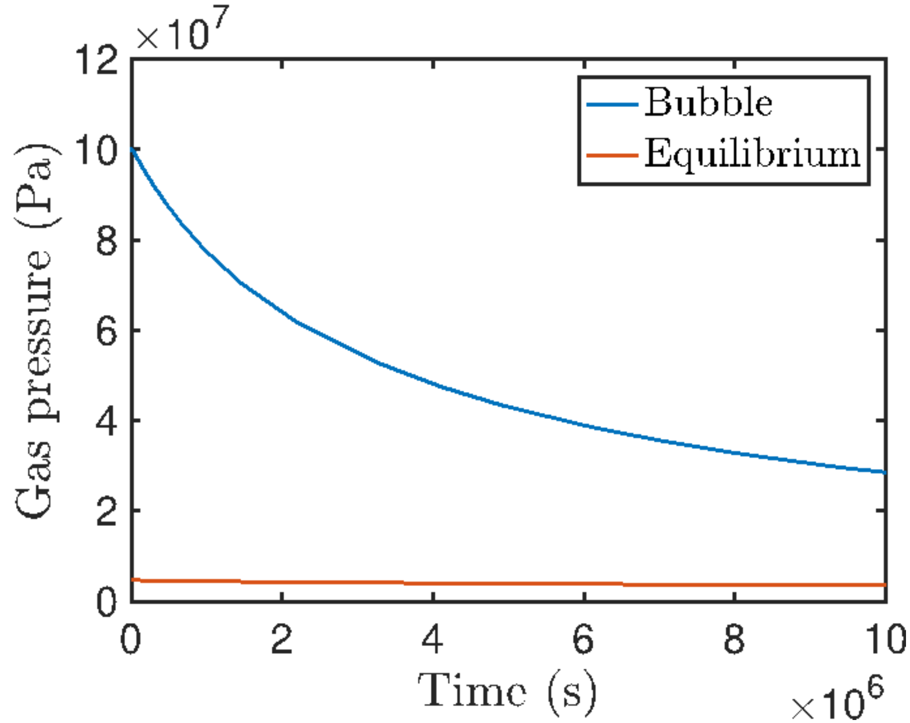
- Considered stress state in equilibrium, underpressurized, overpressurized bubbles (1D simulation in radial coordinates)
  - Equilibrium: σ_{rr} = 0 in surrounding solid matrix
  - Under/overpressurized: σ_{rr} = +/-, corresponding to tensile/compressive stress in surrounding matrix
  - Converge to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



Underpressurized Equilibrium Overpressurized

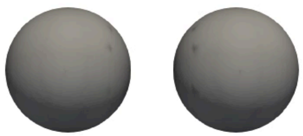
Bubble growth during steady-state operation





- 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

Bubble growth during steady-state operation



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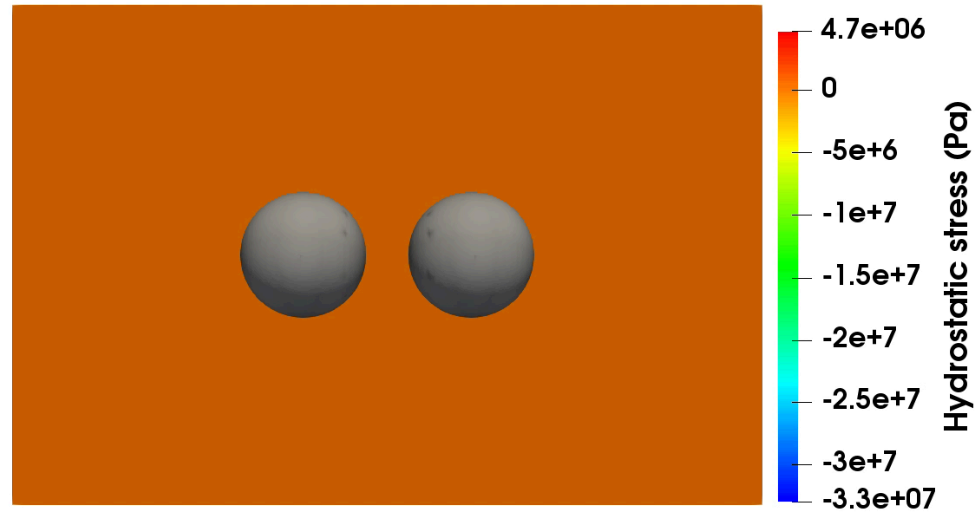
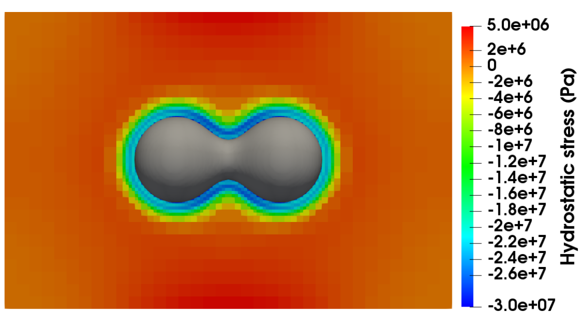


- 3D simulation to 1.5×10^7 s, 2 bubble impingement, initial radii of 300 nm
- Hydrostatic stress surrounding bubbles
  - Region of enhanced compressive hydrostatic stress in “neck”



$t = 1.3 \times 10^6$ s

Bubble response during LOCA transient

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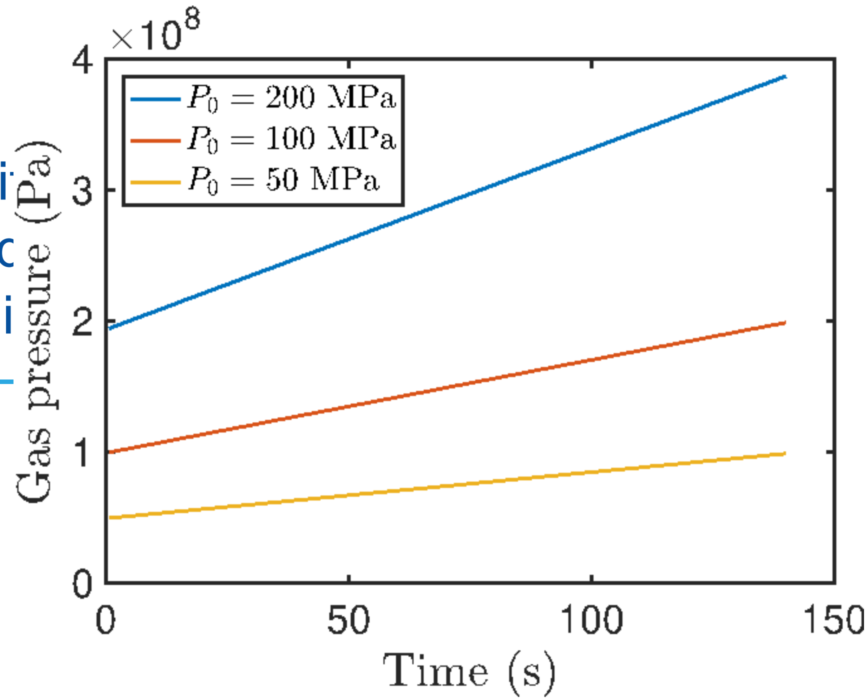

- Temperature ramp from 700 to 1400 K at 5 K/s
- Consider fixed bubble radius of 250 nm in initial conditions
- Maximum initial pressure set to P_0 = 200 MPa (upper bound based on dislocation punching); vary P_0 for fixed bubble size
- No significant change in bubble radius for each case

Bubble response during LOCA transient

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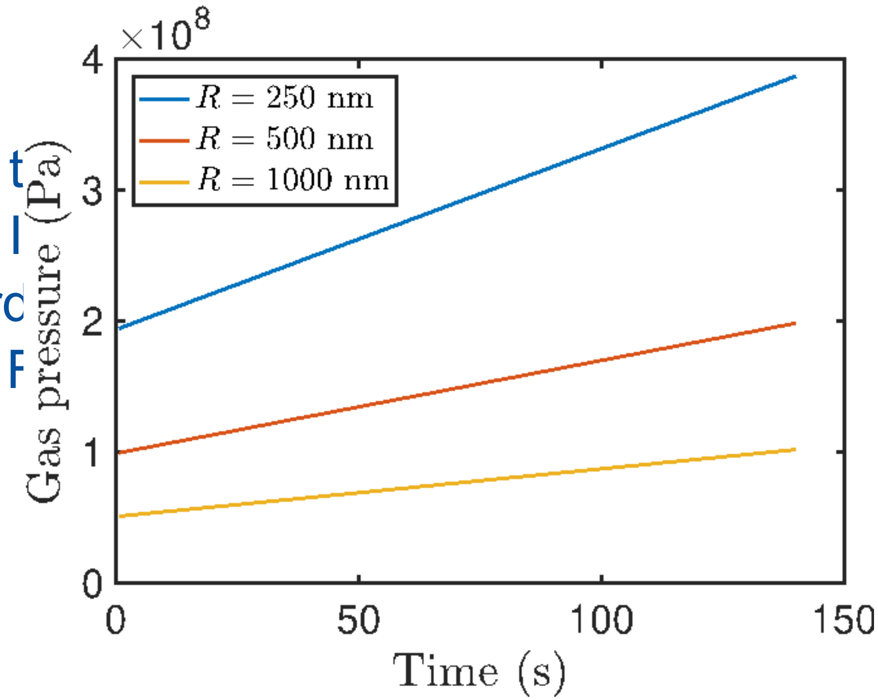

- Vary initial radius: 250 nm, 500 nm, 1000 nm
  - Change domain size to maintain 10% porosity
- Initial pressures set at upper bound estimate from dislocation punching: 200, 100, 50 MPa.
- No significant change in bubble size

Bubble response during LOCA transient

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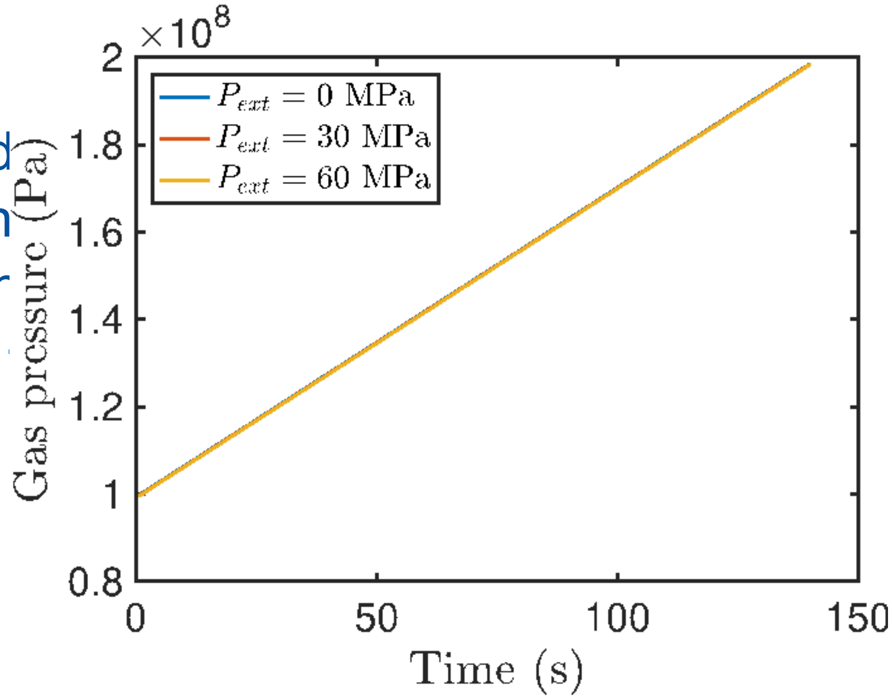
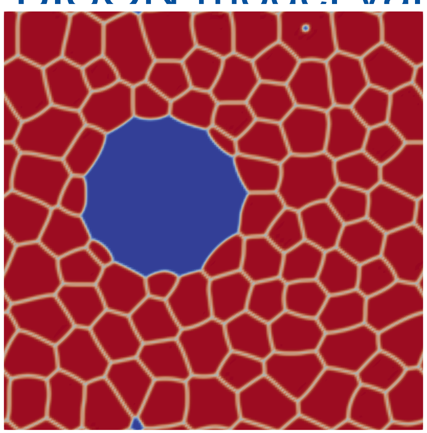


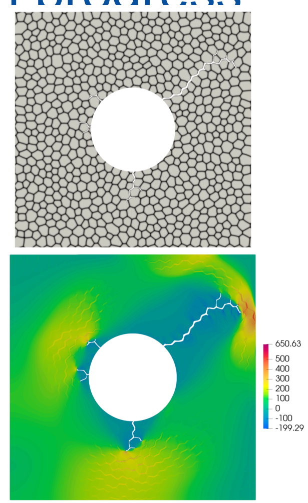
- Vary external pressure at simulation domain boundary, P_{ext}, for constant bubble R = 500 and P_0 = 100 MPa
- No significant size change; pressure transient unchanged
- P_{ext} may have a stronger impact on fracture behavior
- **Based on these results, do not need to consider bubble size change in phase-field fracture model**

Mechanistic Criterion for pulverization of high burnup UO_2 fuel during LOCA

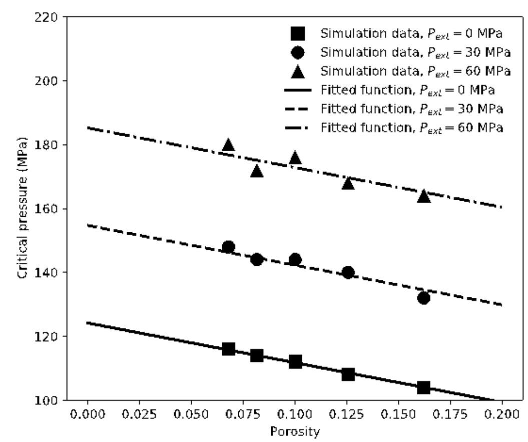
- Estimate pre-LOCA HBS bubble pressure using grand potential-based, multi-grain phase-field model
  - Initial pressure input to BISON
- Phase-field fracture simulations
  - Fit to get BISON criterion as a function of porosity, external stress
- BISON model validation in progress



Bubble Evolution & Pressure



Fracture during LOCA



Bison Criterion

Conclusions: High-burnup UO_2 response to LOCA transients

- Developed new phase-field model that accounts for effects of surface tension and gas bubble pressure to understand non-equilibrium bubbles
- Bubble size did not change significantly during LOCA transients
- Pressure as a function of time determined for given transients
  - Lack of bubble size change allows linear pressure increase with temperature in phase-field fracture model
- Initial implementation of BISON pulverization model completed, validation in progress

Thank you!

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Questions?

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