INL/CON-22-69321-Revision-0

Multi-scale simulation of high burnup UO2 nuclear fuel during loss-of-coolant accident conditions

October 2022

Changing the World's Energy Future

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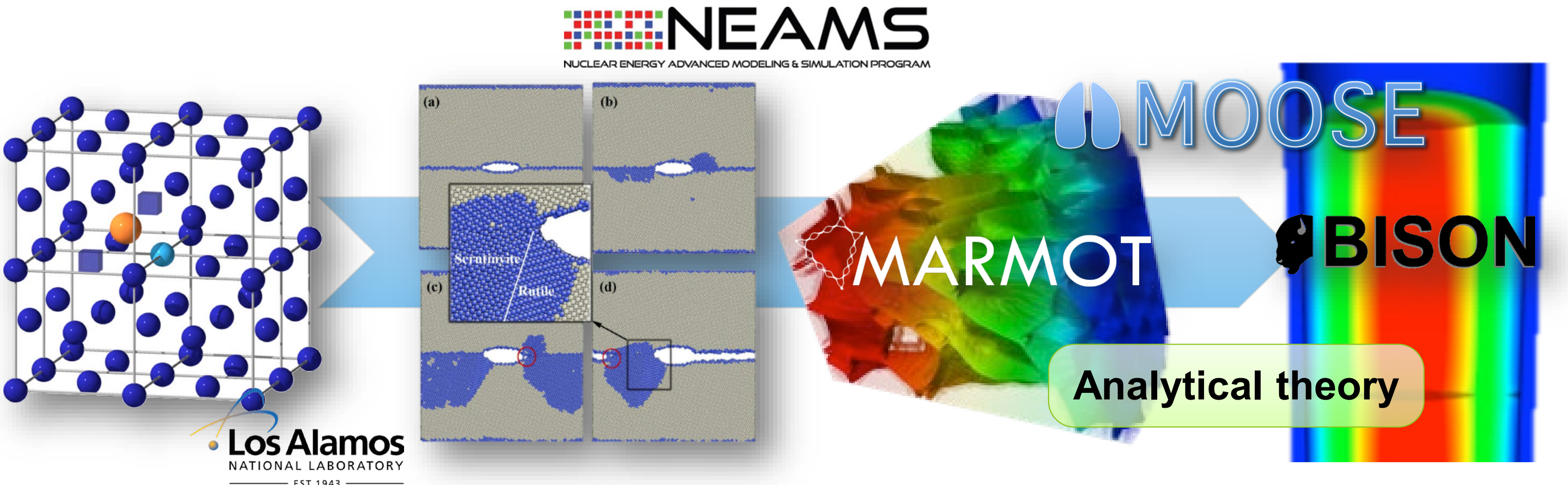
Multiscale fuel performance simulation

- 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

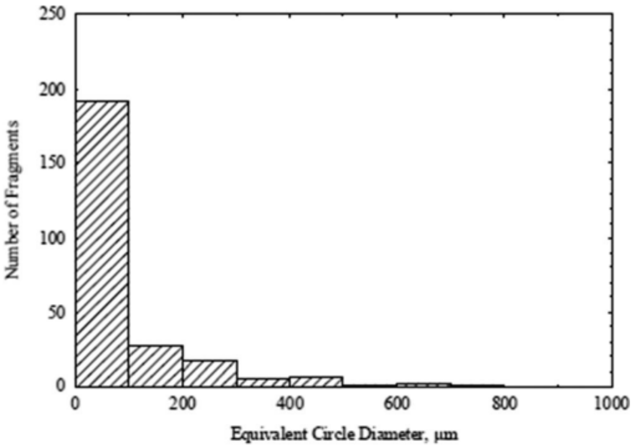
100's of nanometers
Molecular Dynamics
- Identify interfacial mechanisms
- Determine interfacial properties

microns
Mesoscale
- Predict microstructure evolution
- Determine impact on properties

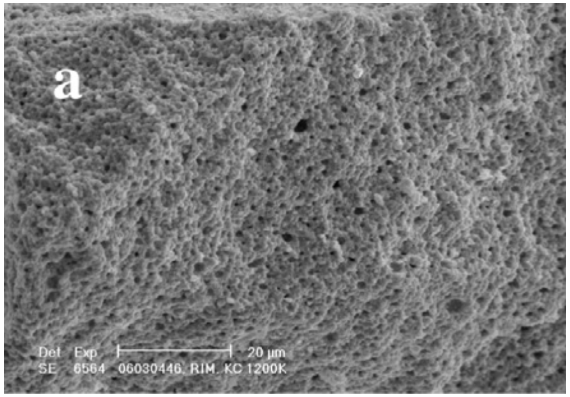
millimeters and up
Engineering Scale
- Use analytical theory
- Predict fuel performance

Fine fragmentation/pulverization of high-burnup UO_2

- 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



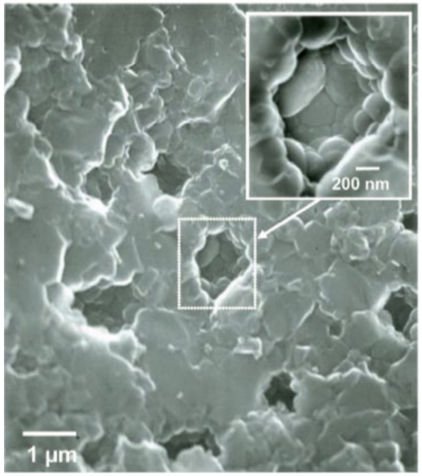
Turnbull et al., Nuc. Sci. & Eng., 179,477 (2015).



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

BISON model for pulverization

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

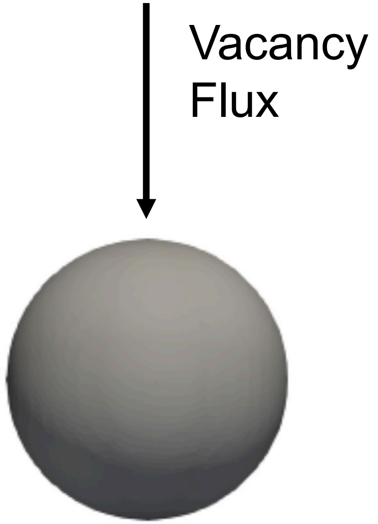


High-burnup structure in UO2 [Sonoda et al., NIMB, 2002].

- 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

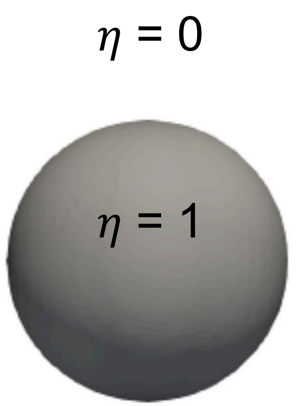
HBS bubble response to LOCA transient

- 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?**



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



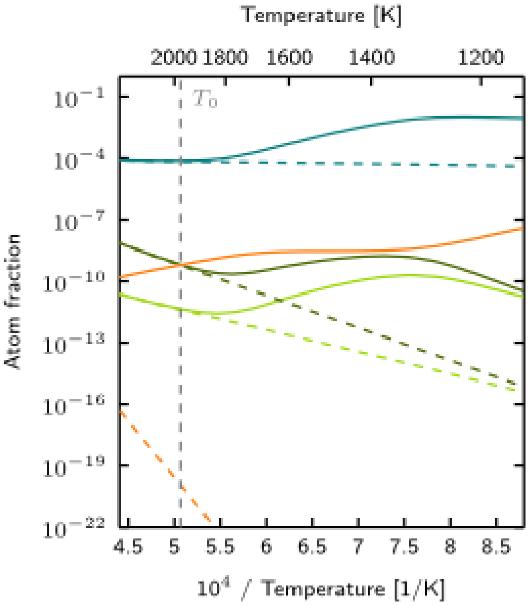
Evolution equations

- 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):

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



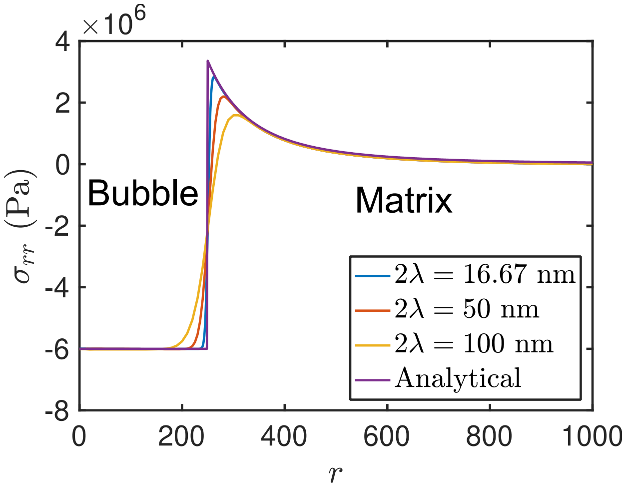
- Mobilities are a function of defect diffusivities:

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

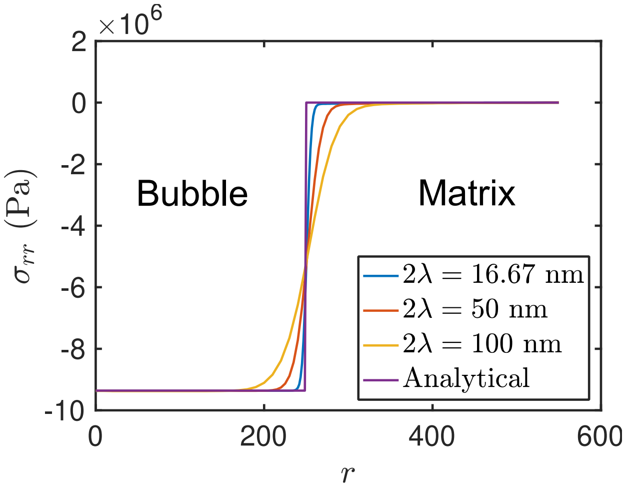
- + KKS system constraints

Phase-field model verification and testing

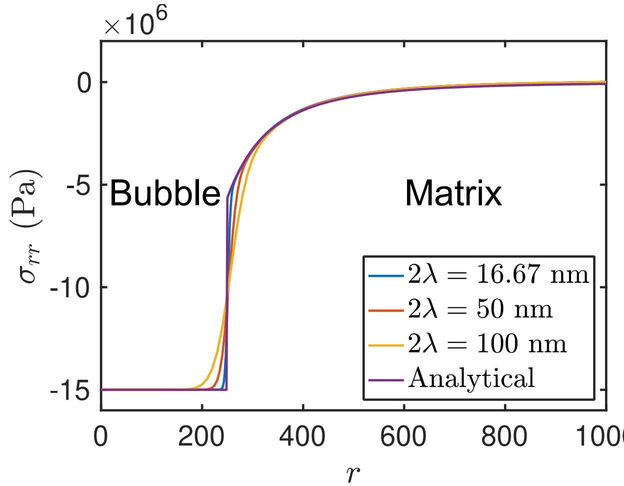
- Considered stress state in equilibrium, underpressurized, overpressurized bubbles (1D simulation in radial coordinates)
  - Equilibrium: σ_{rr} = 0 in surrounding solid matrix
  - Under/overpressurized: σ_{rr} = +/-, corresponding to tensile/compressive stress in surrounding matrix
  - Converge to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



Underpressurized

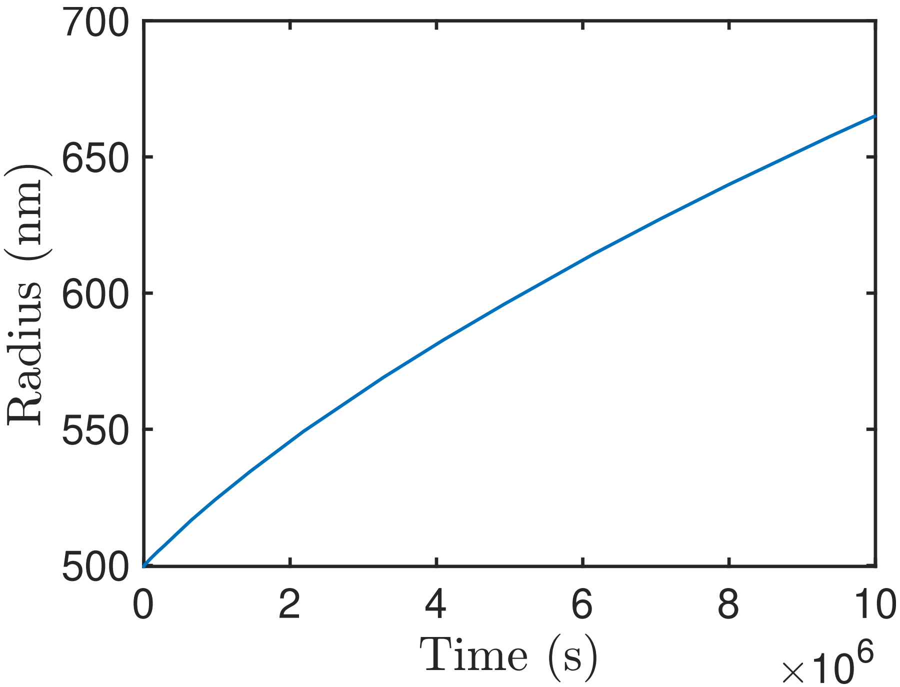


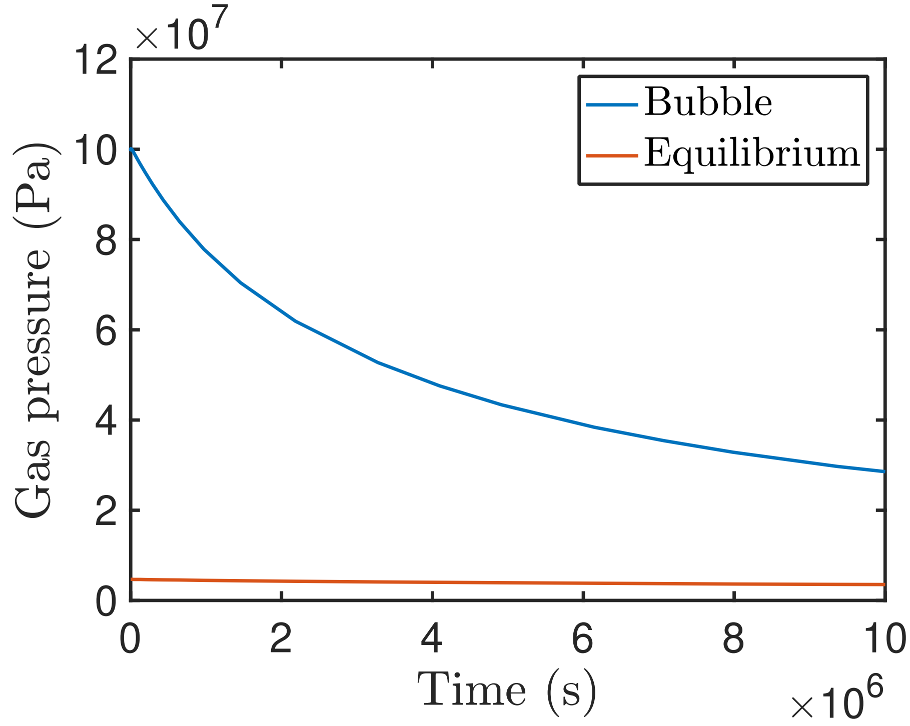
Equilibrium



Overpressurized

Bubble growth during steady-state operation

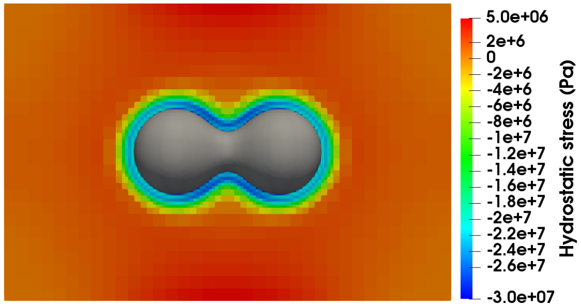




- Assume bubble pressure is 100 MPa in initial conditions
  - Upper bound based on dislocation punching pressure
- Bubble pressure decreases during growth but remains well above equilibrium pressure
  - Increased likelihood of fragmentation during LOCA

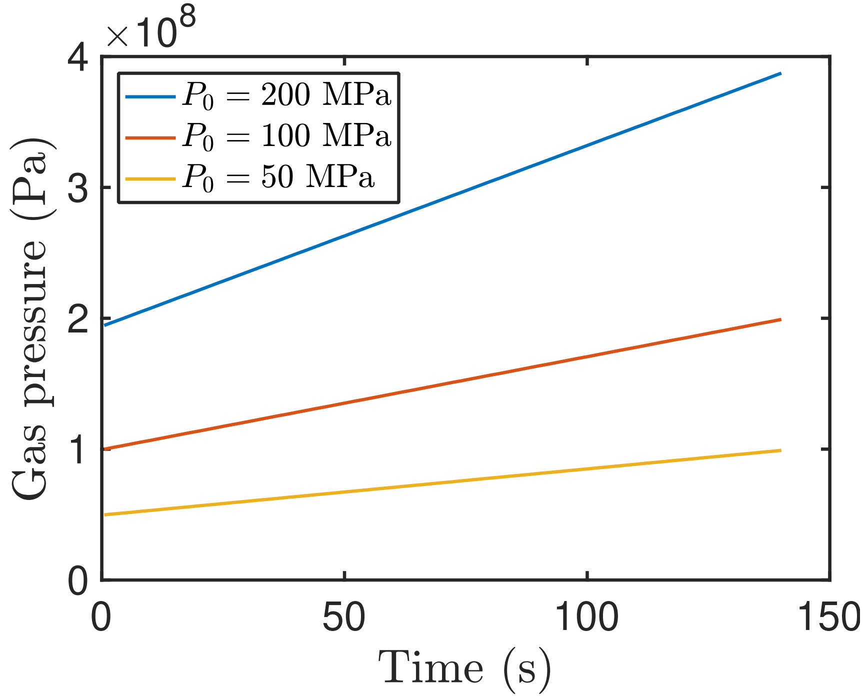
Bubble growth during steady-state operation

- 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”



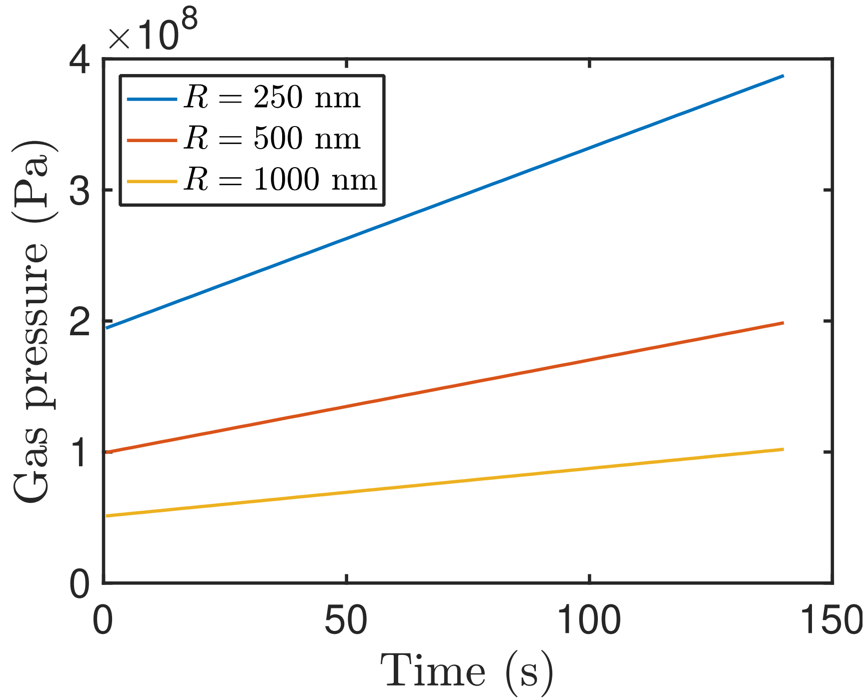
$t = 1.3 \times 10^6$ s

Bubble response during LOCA transient



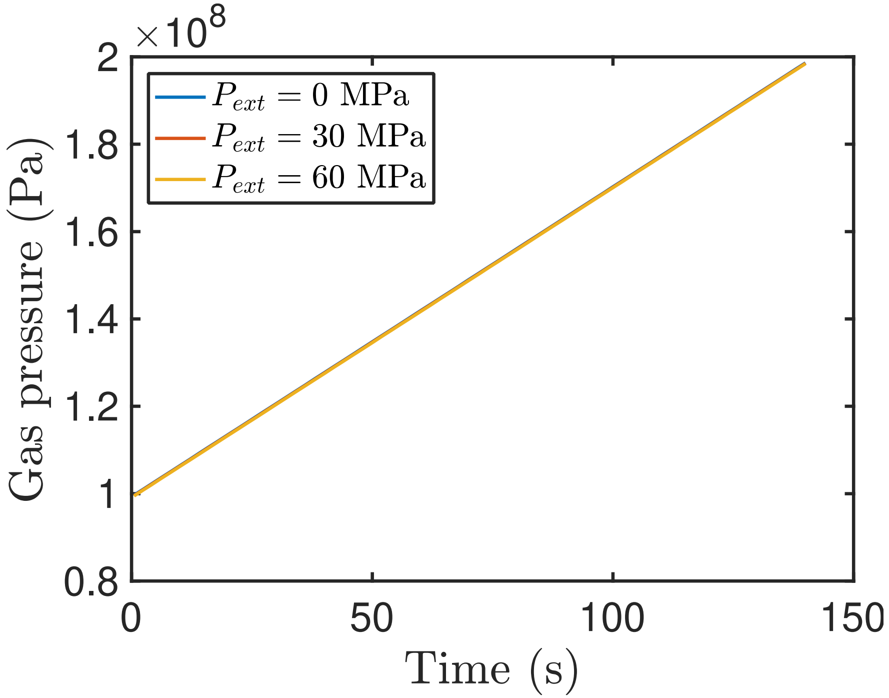
- Temperature ramp from 700 to 1400 K at 5 K/s
- Consider fixed bubble radius of 250 nm in initial conditions
- Maximum initial pressure set to P_0 = 200 MPa (upper bound based on dislocation punching); vary P_0 for fixed bubble size
- No significant change in bubble radius for each case

Bubble response during LOCA transient



- 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

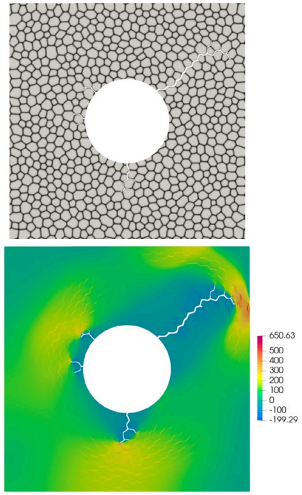
Bubble response during LOCA transient

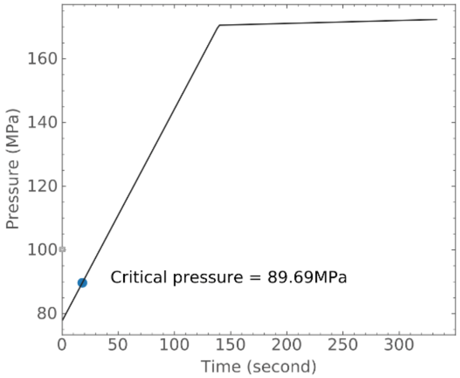


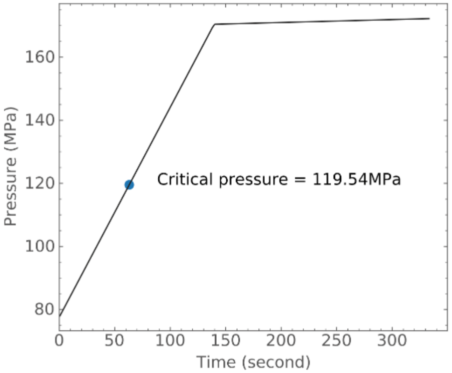
- Vary external pressure at simulation domain boundary, P_{ext}, for constant bubble R = 500 and P_0 = 100 MPa
- No significant size change; pressure transient unchanged
- P_{ext} may have a stronger impact on fracture behavior
- **Based on these results, do not need to consider bubble size change in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

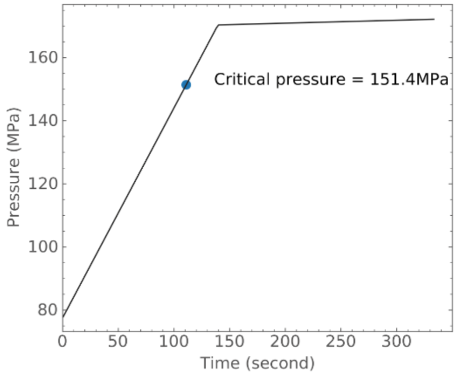
Phase-field fracture modeling to determine pulverization criterion for high burnup UO_2

- Quasi-brittle phase-field fracture model. Essential physics:
  - Order parameter represents crack. HBS grain structure in 2D.
  - Critical fracture strength independent of length scale
  - Internal bubble pressure and external stress at boundaries
- Simulate LOCA transient by ramping bubble internal pressure at rate corresponding to 5K/s temperature transient ($r = 0.5\ \mu$m)
  - Determine critical pressure at which crack formation begins





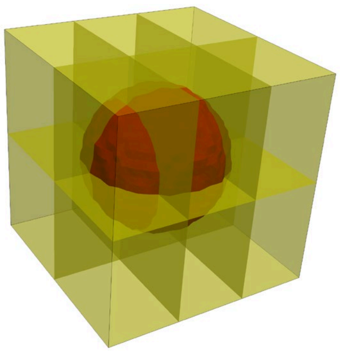


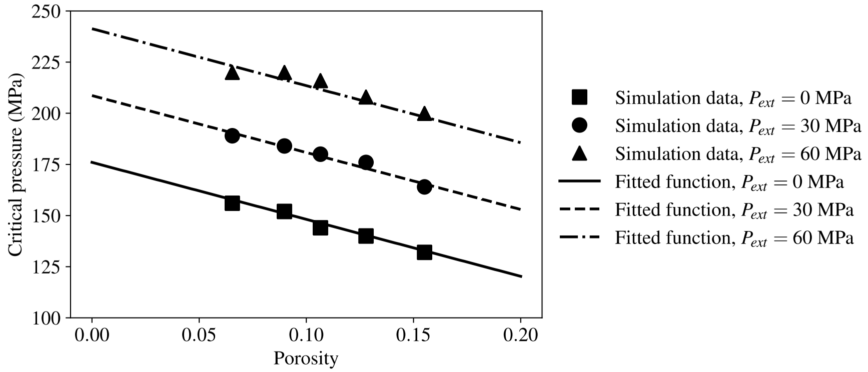


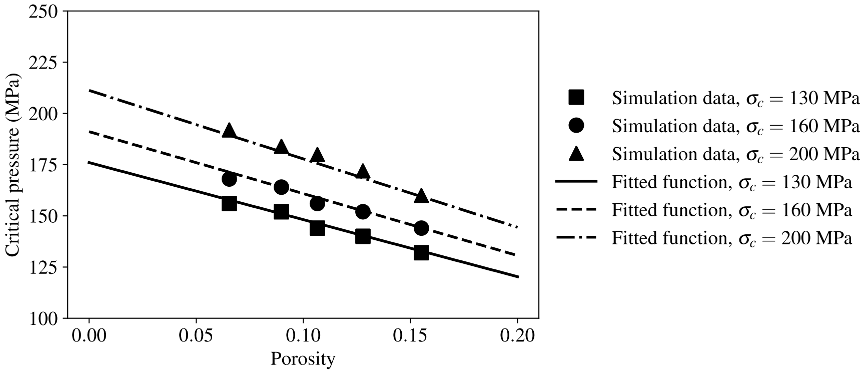
Jiang et al., Theor. Appl. Fract. Mech., 119, 103349 (2022)

Phase-field fracture modeling to determine pulverization criterion for high burnup UO_2

- Did 3D simulations to get critical pressure for crack initiation, use as pulverization criterion
  - Varied external pressure, GB fracture strength. Maintained constant bubble size ($r = 0.5\ \mu$m), varied domain size to vary porosity
  - 2D simulations showed HBS grain structure didn't change critical pressure significantly, so didn't resolve grain structure in 3D
  - Fit function to data, implemented in BISON







Bison Criterion

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.

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

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

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
- Used phase-field fracture model to simulate crack initiation in High Burnup UO2
  - 3D simulations used to determine pulverization criterion for engineering-scale model
- Comparison to existing empirical threshold and Studsvik Rods 191 and 196

Thank you!

Funding Support: DOE-NE NEAMS Program

NEAMS
NUCLEAR ENERGY ADVANCED MODELING & SIMULATION PROGRAM

Questions?

Phase-field model: Free energy functional

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

- f_{chem} = bulk chemical free energy density. $h(\eta)$ is a smooth interpolation function:

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

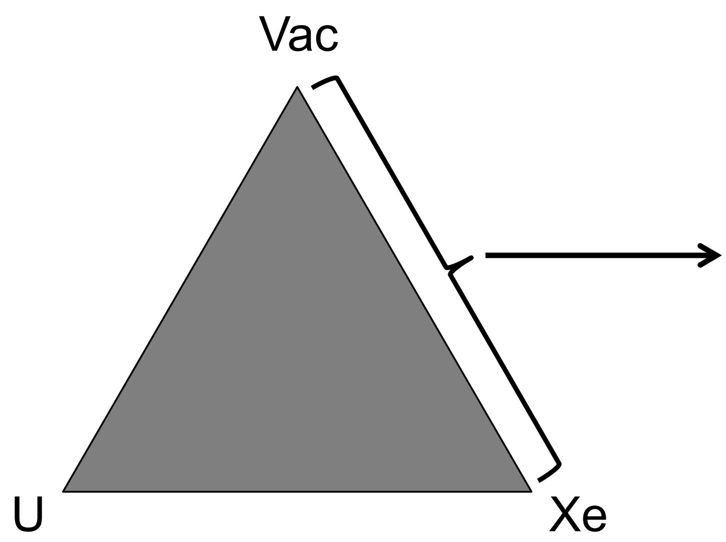
- f^m_{chem} = chemical free energy of the matrix phase. Fit a parabolic approximation to ideal solution energetics:

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

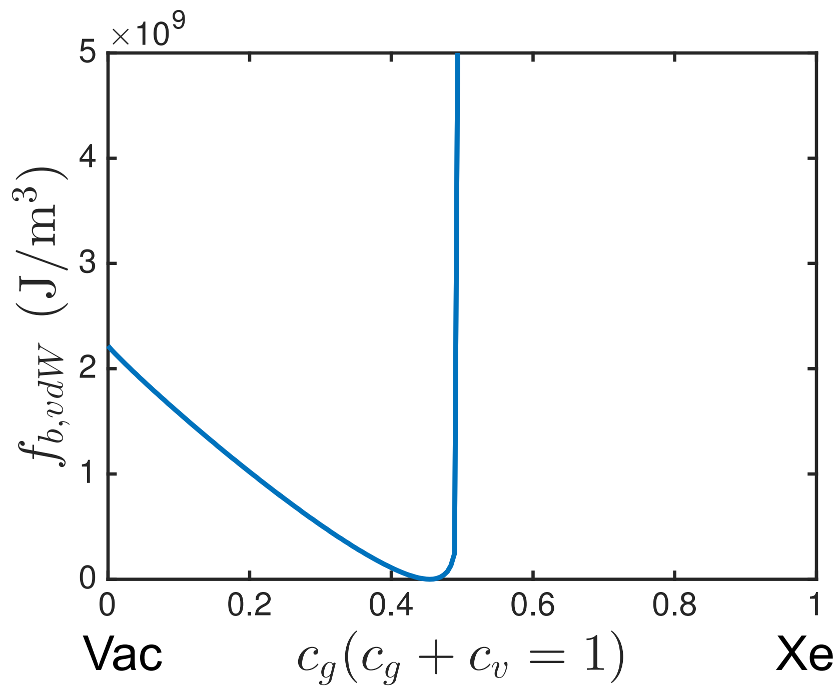
- f^b_{chem} = chemical free energy of the bubble phase. The bubble is considered to be a mixture of gas atoms and U-site vacancies. Energy given by the Helmholtz free energy of a van der Waals gas:

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

Parameterization for Xe gas phase



Gibbs triangle: U lattice sites



Helmholtz free energy density: Van der Waals gas

Phase-field model: Elastic energy

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

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

- Mechanical equilibrium equation:

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

- Eigenstrain due to vacancies:

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

- Bubble pressure:

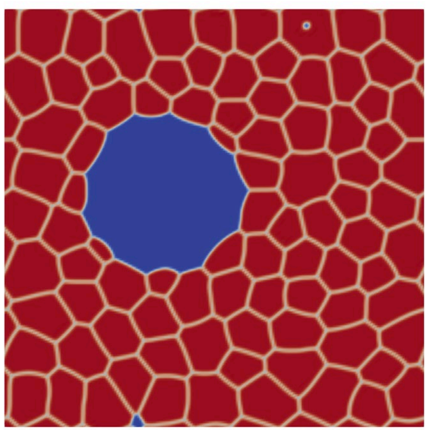
$$\sigma_{ij}^b = -\left(\frac{kT}{\frac{V_a}{c_g^b} - b} \right) \mathbf{I} + C_{ijkl}^b \epsilon_{kl}^{el,b}$$

- Surface tension:

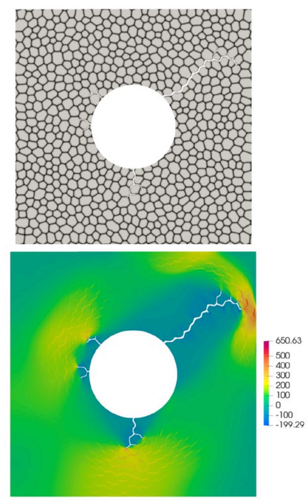
$$\sigma_{ij}^{st} = \left[W g(\eta) + \frac{\kappa}{2} |\nabla \eta|^2 \right] \mathbf{I} - \kappa \nabla \eta \otimes \nabla \eta$$

Mechanistic criterion for pulverization of high burnup UO_2 fuel during LOCA

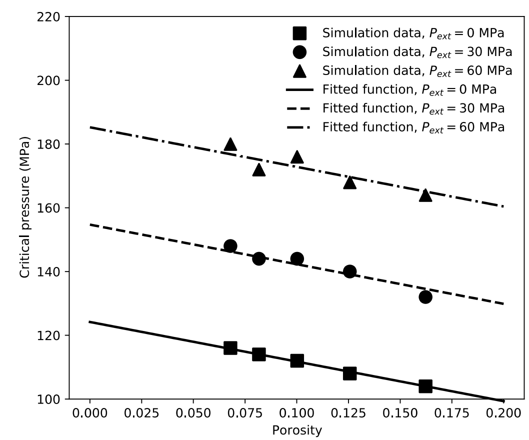
- Estimate pre-LOCA HBS bubble pressure using grand potential-based, multi-grain phase-field model
  - Initial pressure input to BISON
- Phase-field fracture simulations
  - Fit to get BISON criterion as a function of porosity, external stress
- BISON model validation in progress



Bubble Evolution & Pressure



Fracture during LOCA



Bison Criterion