INL/MIS-21-65373-Revision-0

Development of a physics-based pulverization criterion for high burnup UO2 fuel in BISON

December 2021

Changing the World's Energy Future

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Idaho National Laboratory

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Development of a physics-based pulverization criterion for high burnup UO2 fuel in BISON

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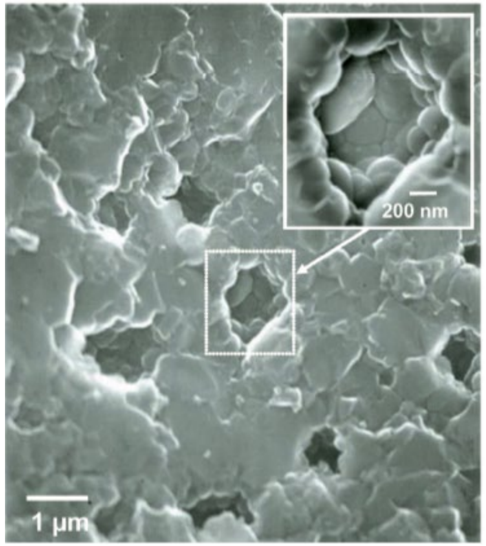
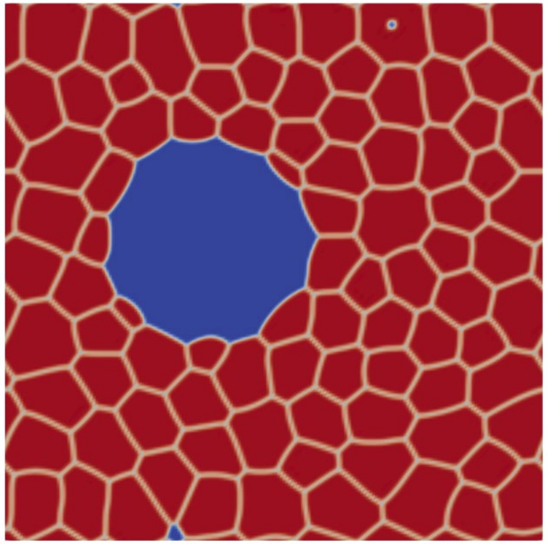
U.S. DEPARTMENT OF ENERGY

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 at that quadrature point
- Mesoscale work to inform BISON calculation of P_g, P_g^{cr}



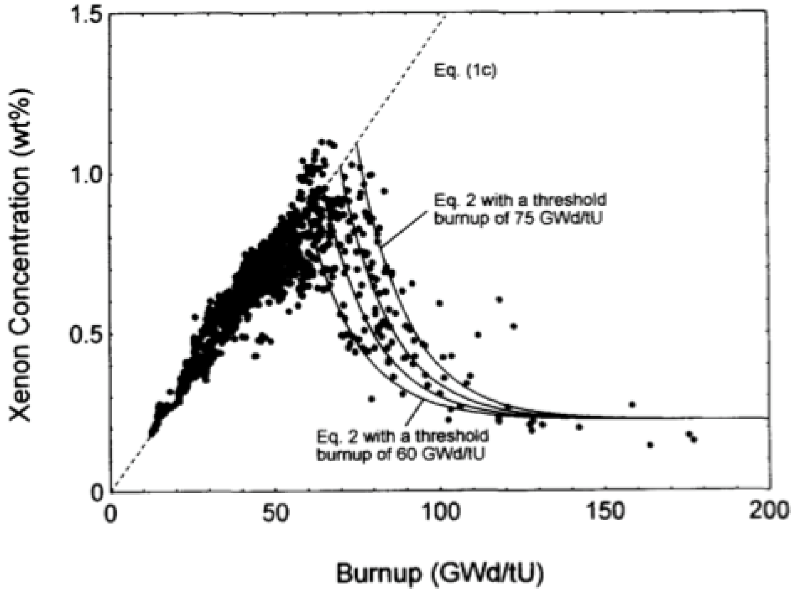


Mesoscale simulations to inform BISON calculation of Bubble Pressure

- BISON SIFGRS model currently used to determine bubble pressure prior to HBS formation
  - Does not account for change in morphology of bubbles upon HBS formation
- 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:

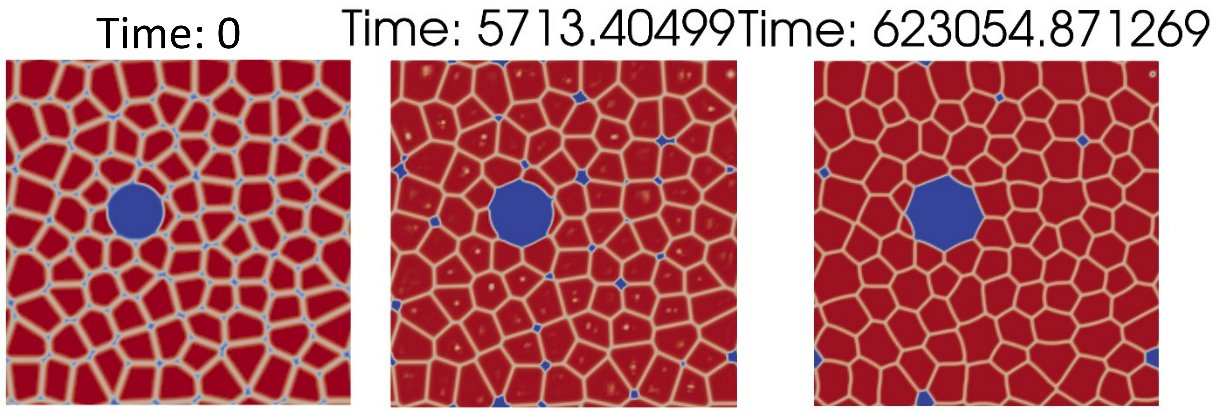
$$P = P_{eq} + P_{ex} = P_H + \frac{2\gamma}{r} + \frac{Gb}{r}$$



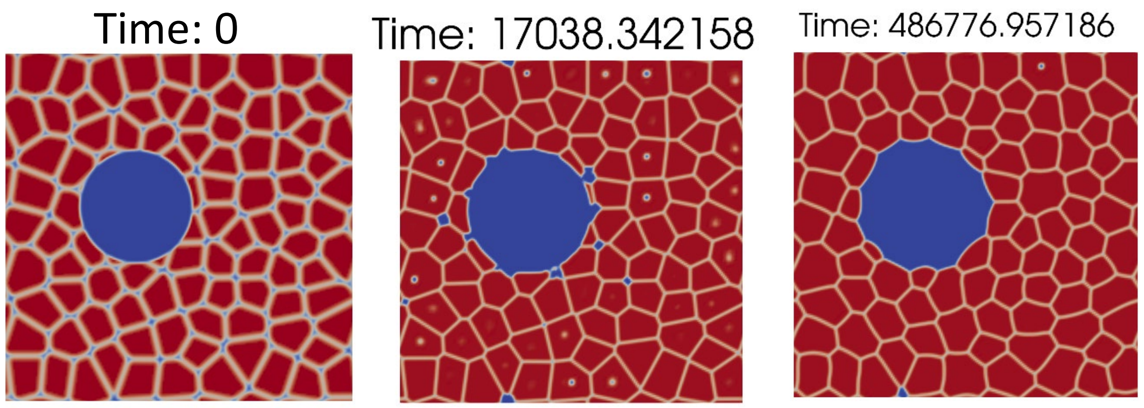
Intragranular Xe concentration
Lassman et al., JNM (1995)

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

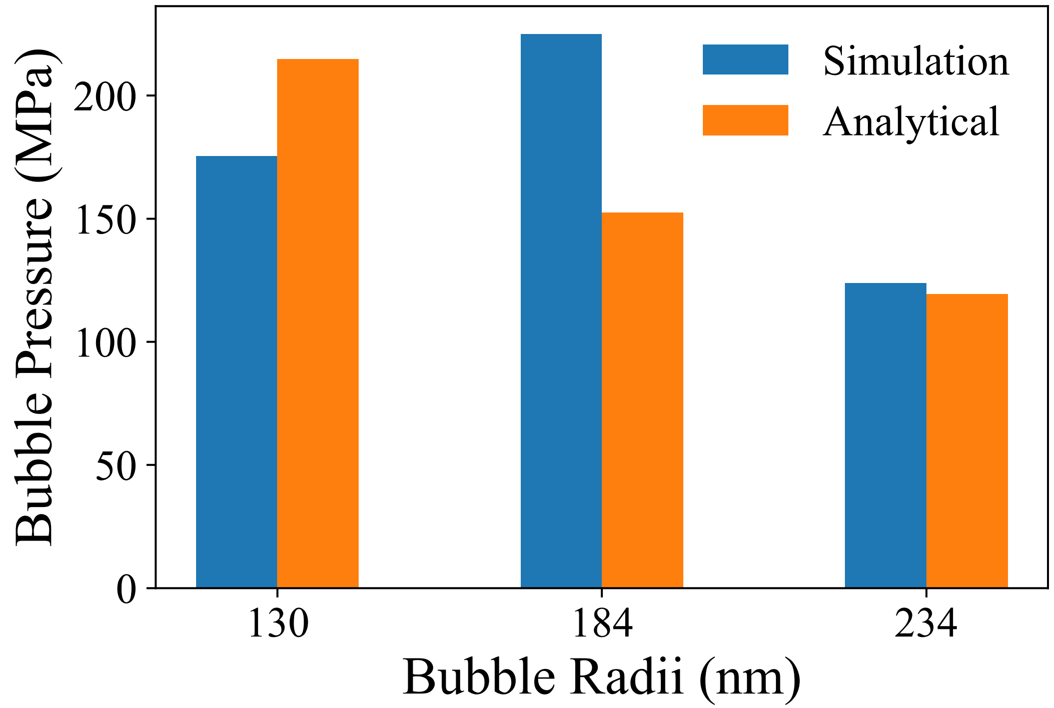


Initial bubble radius: 100 nm



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:

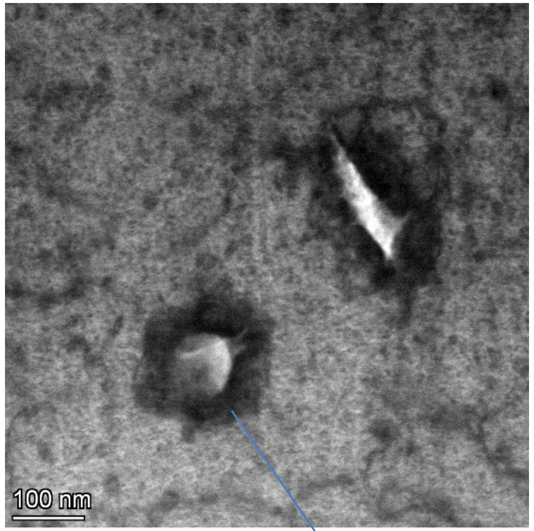
$$P = P_{eq} + P_{ex} = P_H + \frac{2\gamma}{r} + \frac{Gb}{r}$$

- 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**

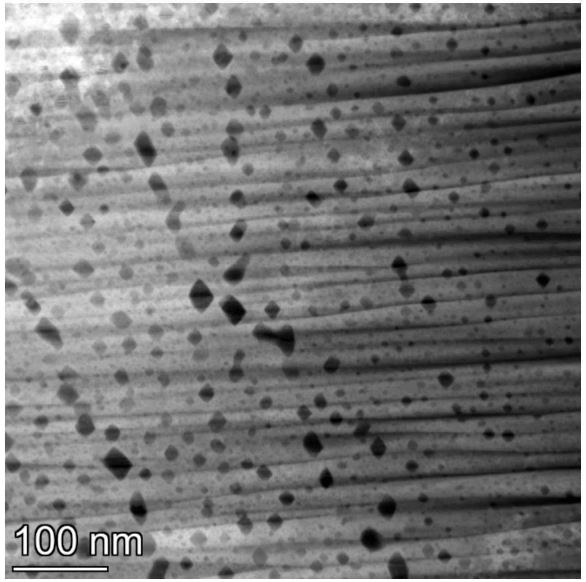
Advanced technique assessment for bubble pressure determination

- (Accurate) experimental determination of bubble pressure in nuclear fuels is one of the most needed and yet most challenging tasks

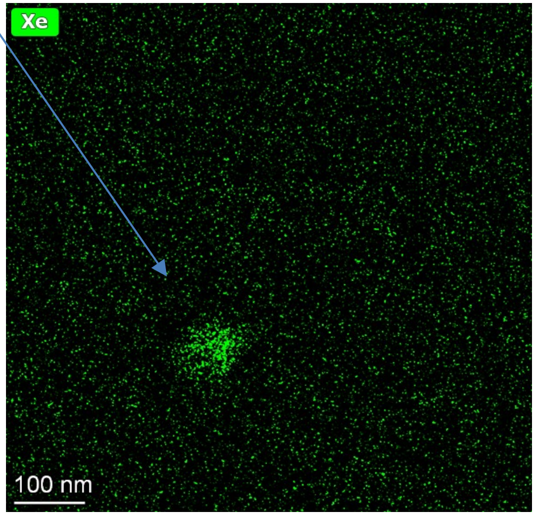
- Proposal awarded to perform 4D STEM and EELS to map the strain around the bubbles at the National Center of Electron Microscopy (NCEM) using a double-aberration-corrected scanning transmission electron microscopy (STEM/TEM)



Matrix deformation around the pressurized bubble in high burnup UO_2 fuel



Intragranular Xe bubbles in ion-implanted UO2 simulating HBS formation



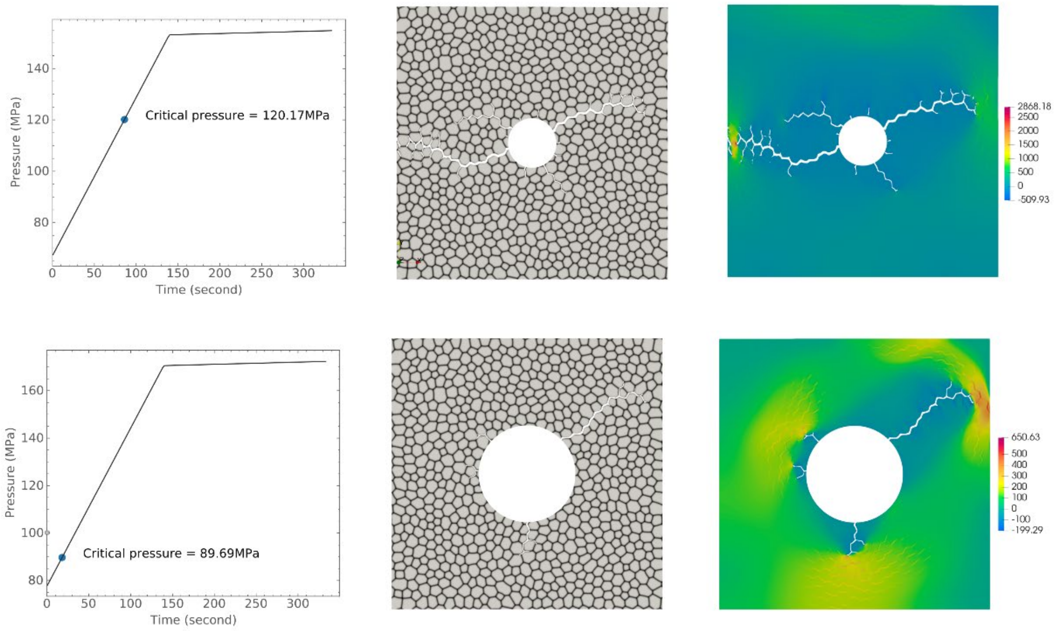
Phase-field fragmentation modeling

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

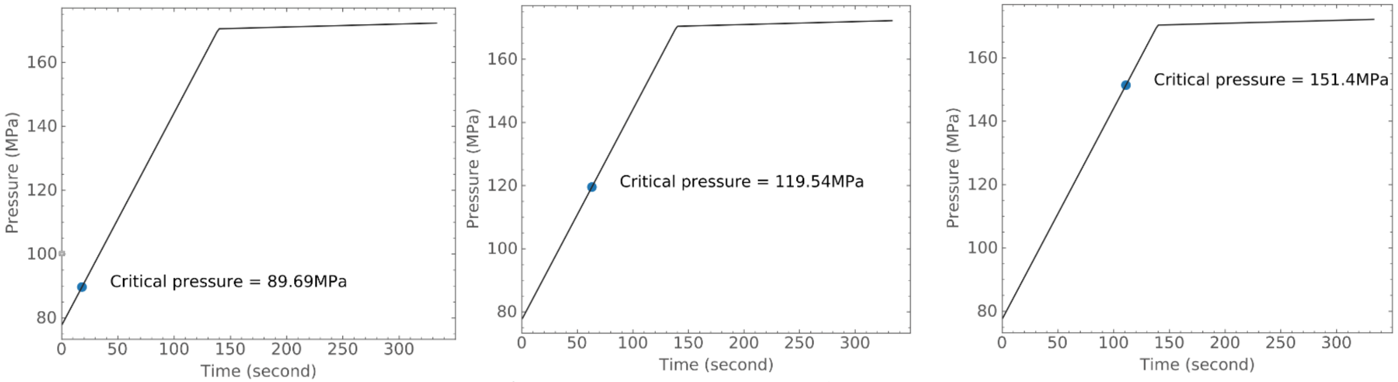
- It remains elastic behavior before crack initiates
- Critical fracture strength is independent of length-scale parameter
- It can predict general softening laws

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

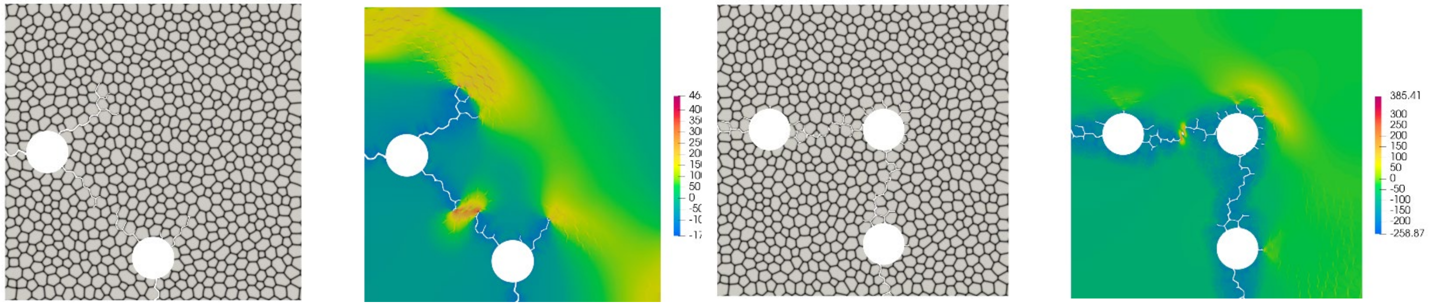
- Effect of bubble sizes: critical pressure is lower for the larger bubble
- Effect of external pressure: critical pressure becomes higher for larger external pressure values
- Effect of bubble interaction: fragmentation size is likely determined by bubble spatial distribution



Bubble size = 0.25 μm and 0.5 μm



External pressure = 0, 30 and 60 MPa

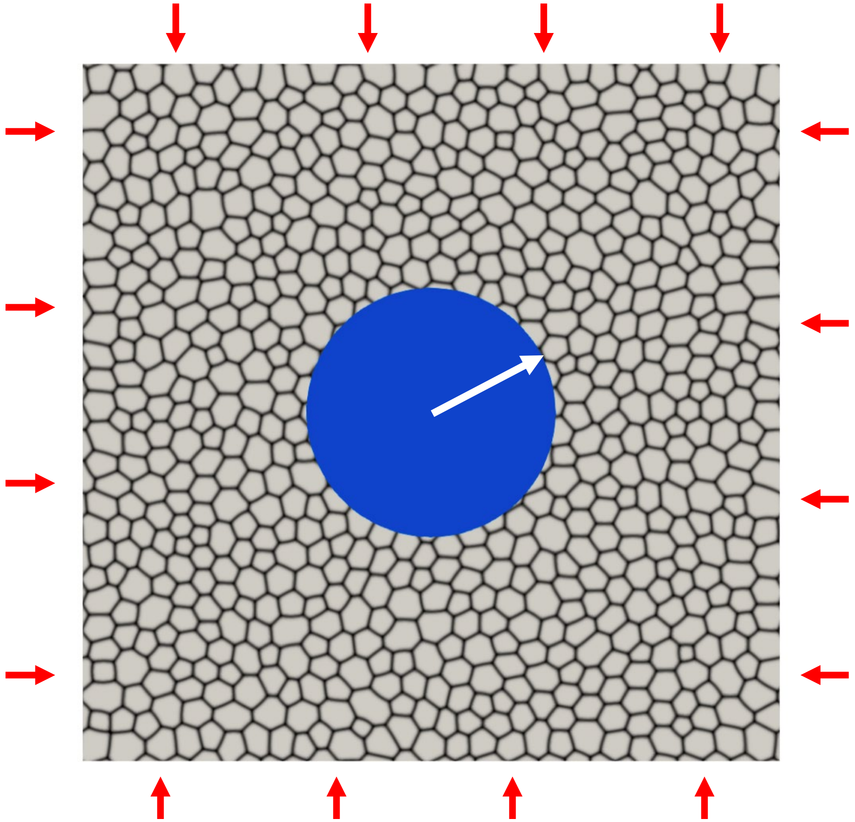


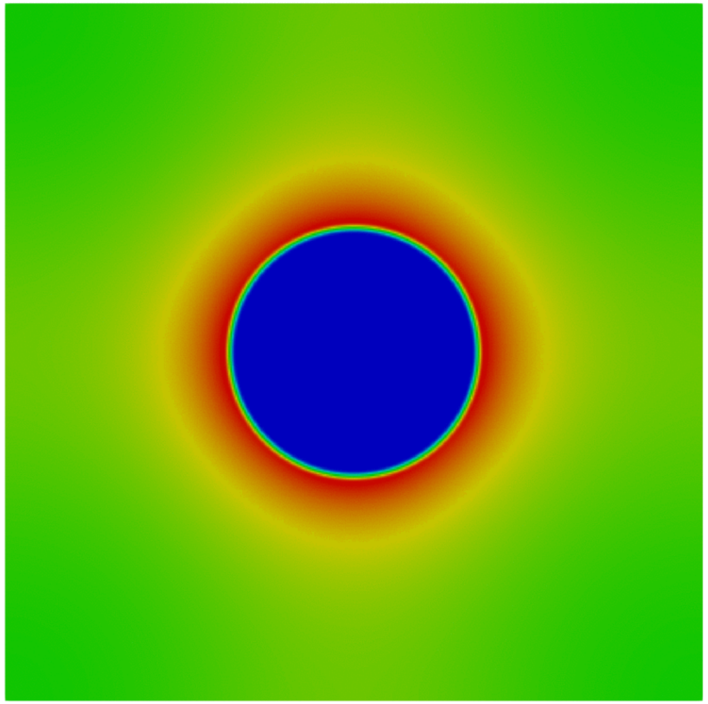
Two and three bubbles interaction

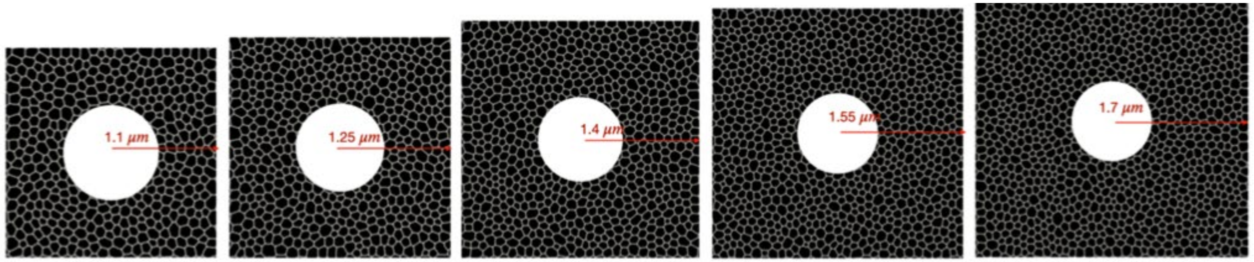
Determine BISON pulverization criterion

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

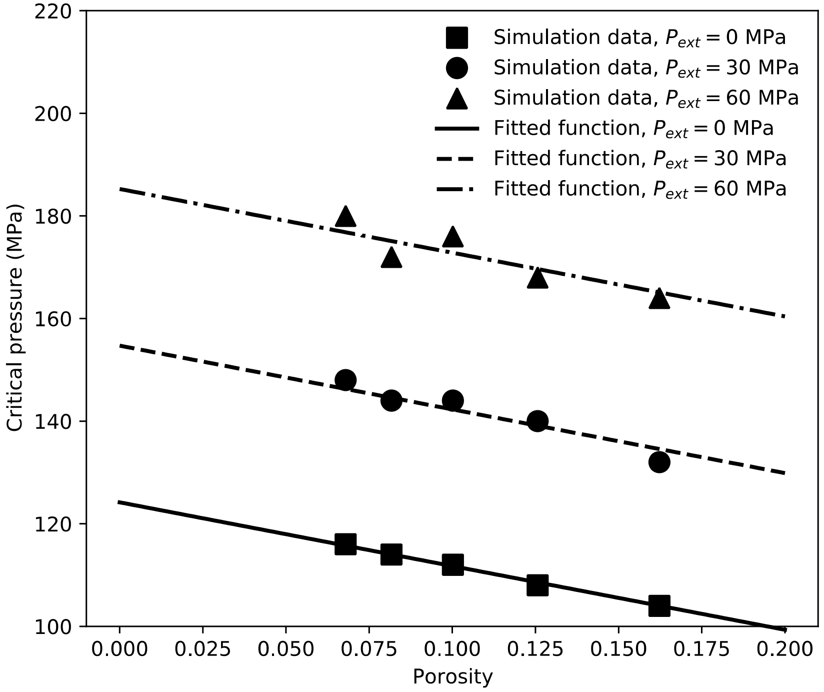
- Use periodic boundary conditions to account for multi-bubble interaction.
- Consider varying porosity (p), external pressure (p_{ext}), critical fracture stress of grain boundary (σ_c)

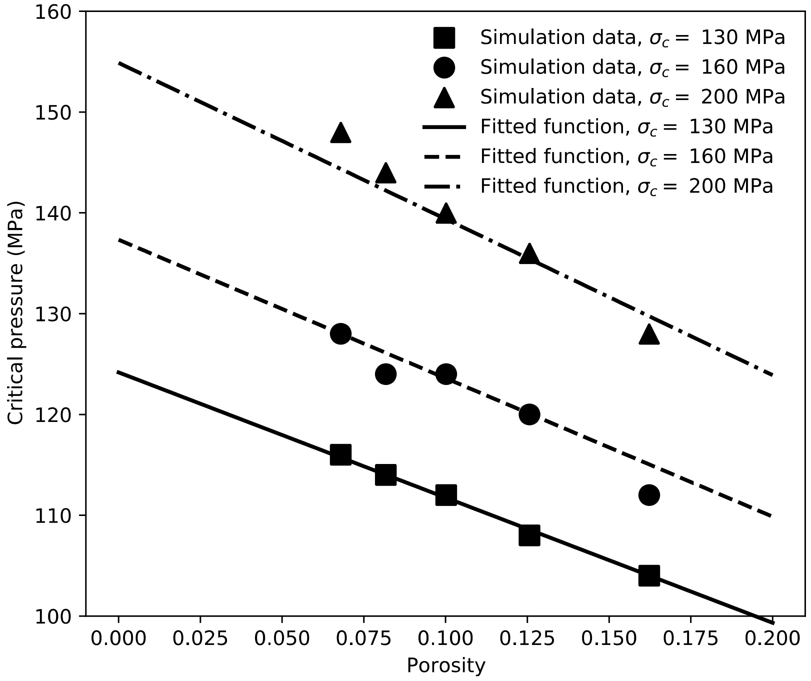






BISON pulverization criterion





- 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, using both analytical and phase-field fracture-based criteria
- Future work:
  - Inform σ_c, G_c from atomistic simulations (LANL) and experiment (AFC)

Multilab M2 Milestone: Studsvik Rods 191 and 196 Revisit

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.

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

First experimental measurement of fracture strength in HBu

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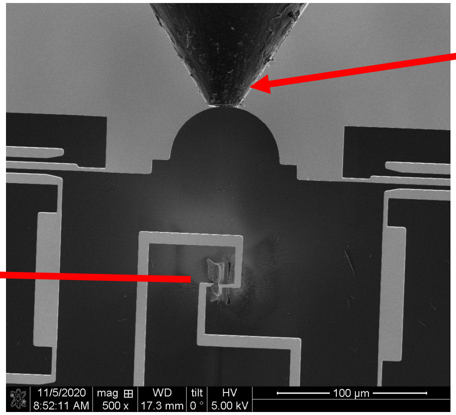
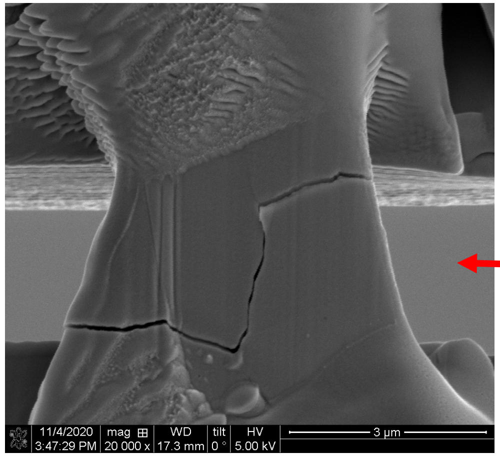


Indenter tip

Micro tensile bar



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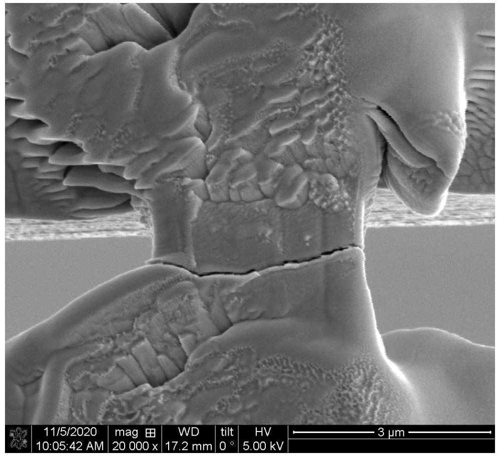

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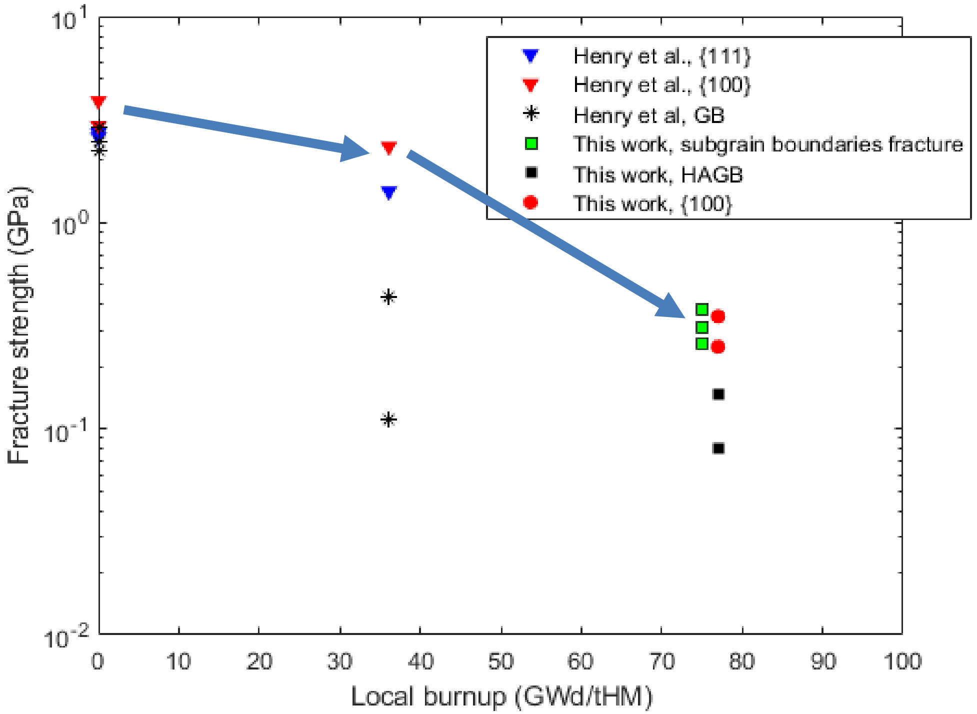
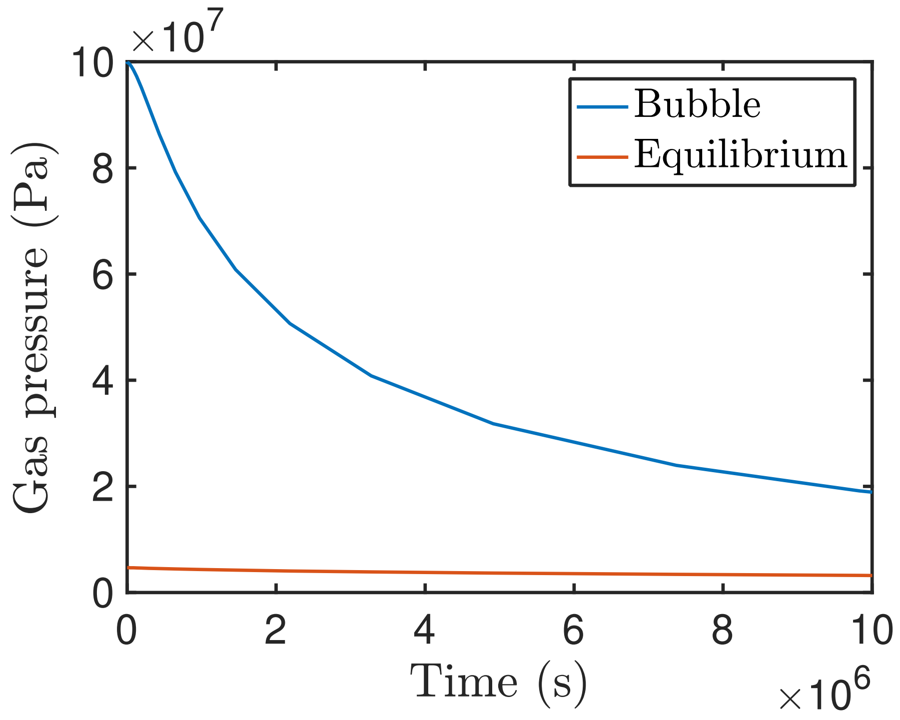


- Primary input for fragmentation criteria
- Leveraging new instrumentation to address the data gap from the critical review from *Capps et al. JNM 546 (2021)*
- Data highlight decrease in both inter- and intra-granular fracture strength

FY22 plans

- Improve estimates of initial HBS bubble pressure at formation
  - Incorporate additional defect species using LANL data
  - Marmot-Xolotl coupling
- Model for HBS bubble pressure evolution (after formation)
  - Based on current phase-field results, assume growth driven by vacancy flux
  - 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)