Simulation of fission gas in uranium oxide nuclear fuel

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U. Tennessee	Brian Wirth	Abdullah Al-Fadhili, Sophie Blondel

In partnership with:

RAPIDS

FASTMATH

Sandia National Laboratories

UF UNIVERSITY of FLORIDA

Argonne NATIONAL LABORATORY

INL

THE UNIVERSITY OF TENNESSEE KNOXVILLE

DEPARTMENT OF NUCLEAR ENGINEERING

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OAK RIDGE National Laboratory

Project web site:
https://collab.cels.anl.gov/display/Fission GasSciDAC2

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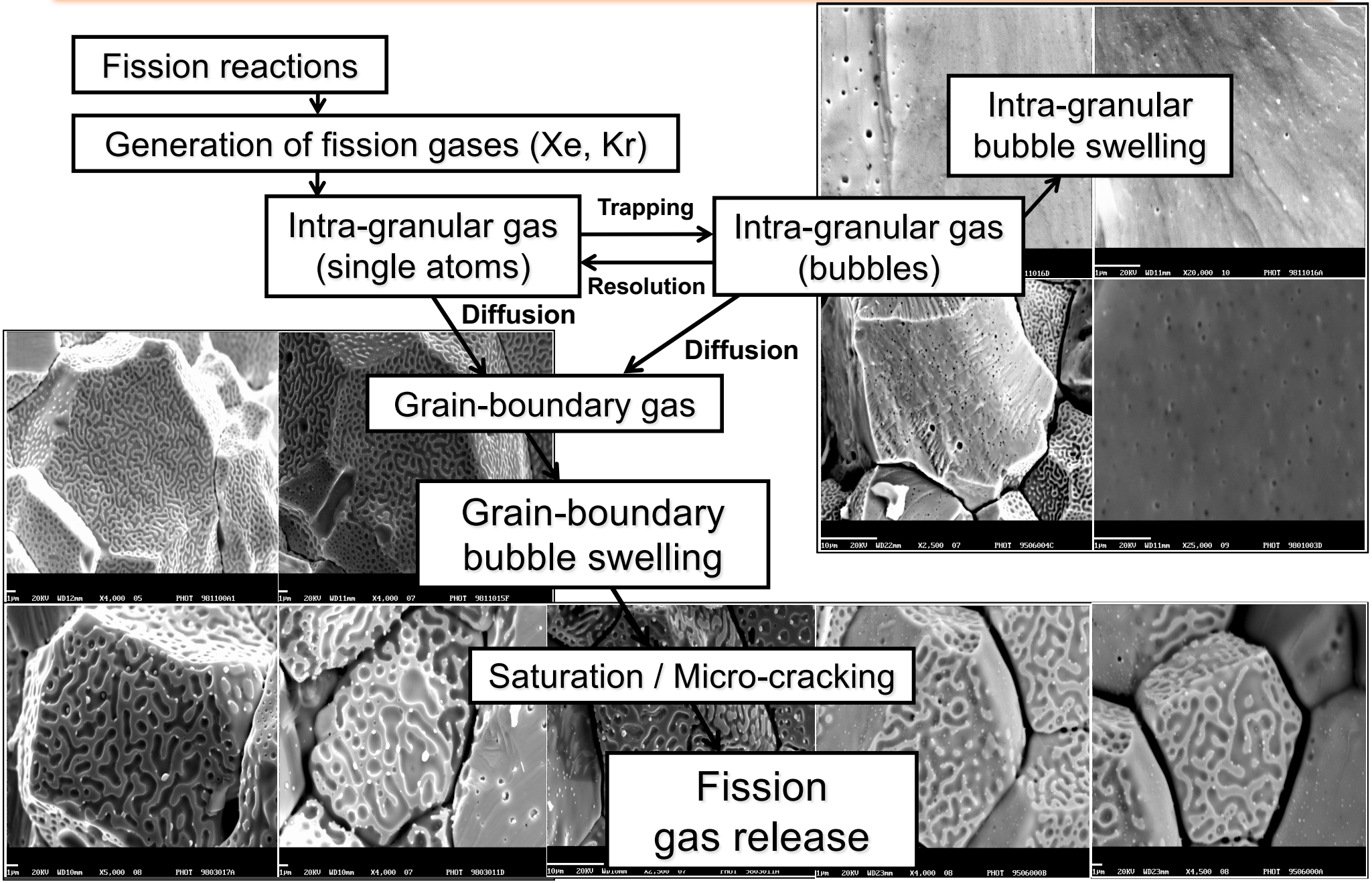
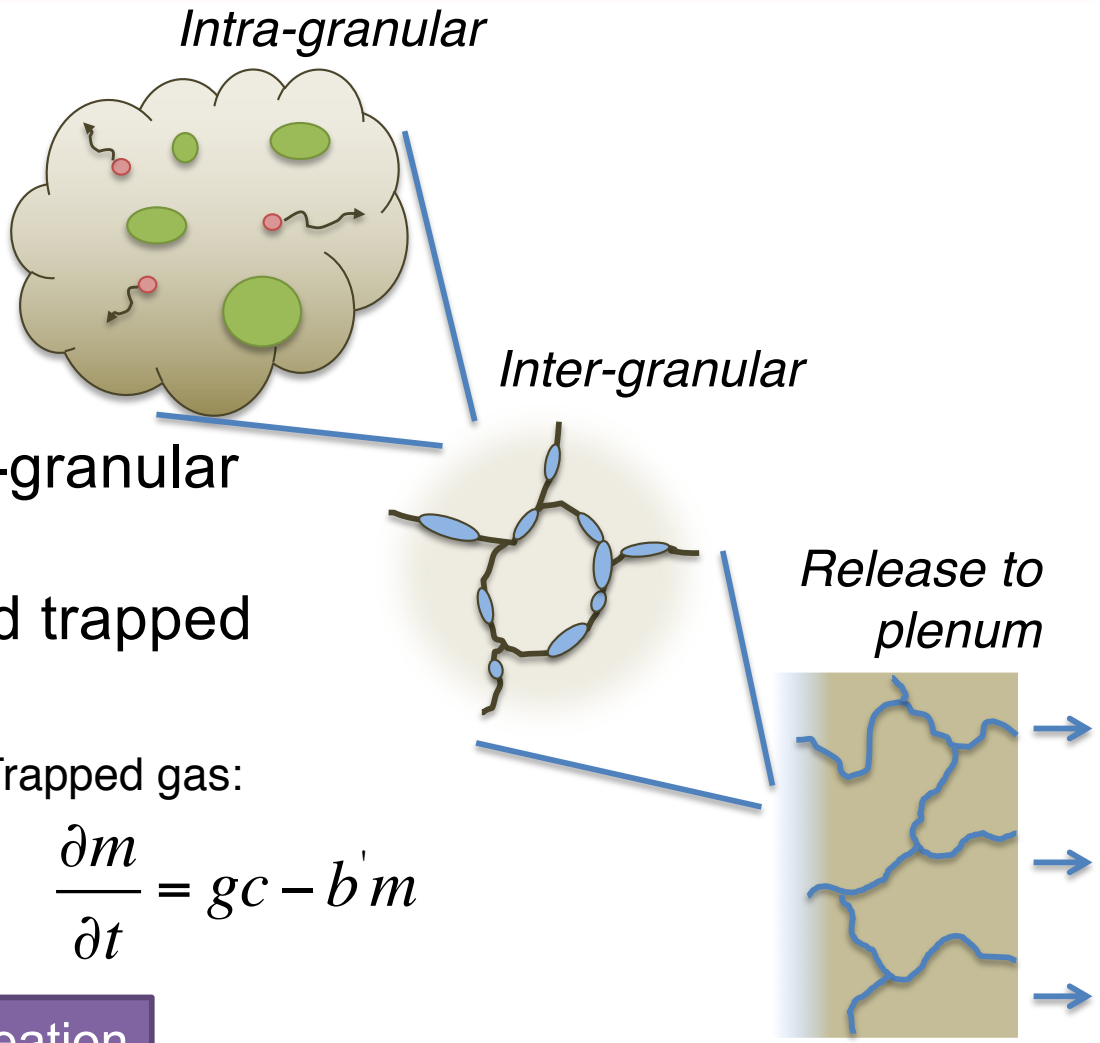
The long-standing fission gas problem



* G. Pastore (INL) – micrographs from White, Corcoran and Barnes, Report R&T/NG/EXT/REP/02060/02 (2006).

The fission gas release process

- Fission gas located:
  - Mobile single gas atoms
  - *Intra*-granular bubbles
  - *Inter*-granular bubbles



- Gas release driven by inter-granular bubble interconnection:
- Concentration of mobile and trapped intragranular gas:

Mobile gas:

$$\frac{\partial c}{\partial t} = D\nabla^2 c - gc + b'm + \dot{\beta}$$

Trapped gas:

$$\frac{\partial m}{\partial t} = gc - b'm$$

Diffusion

Absorption

Re-solution

Creation

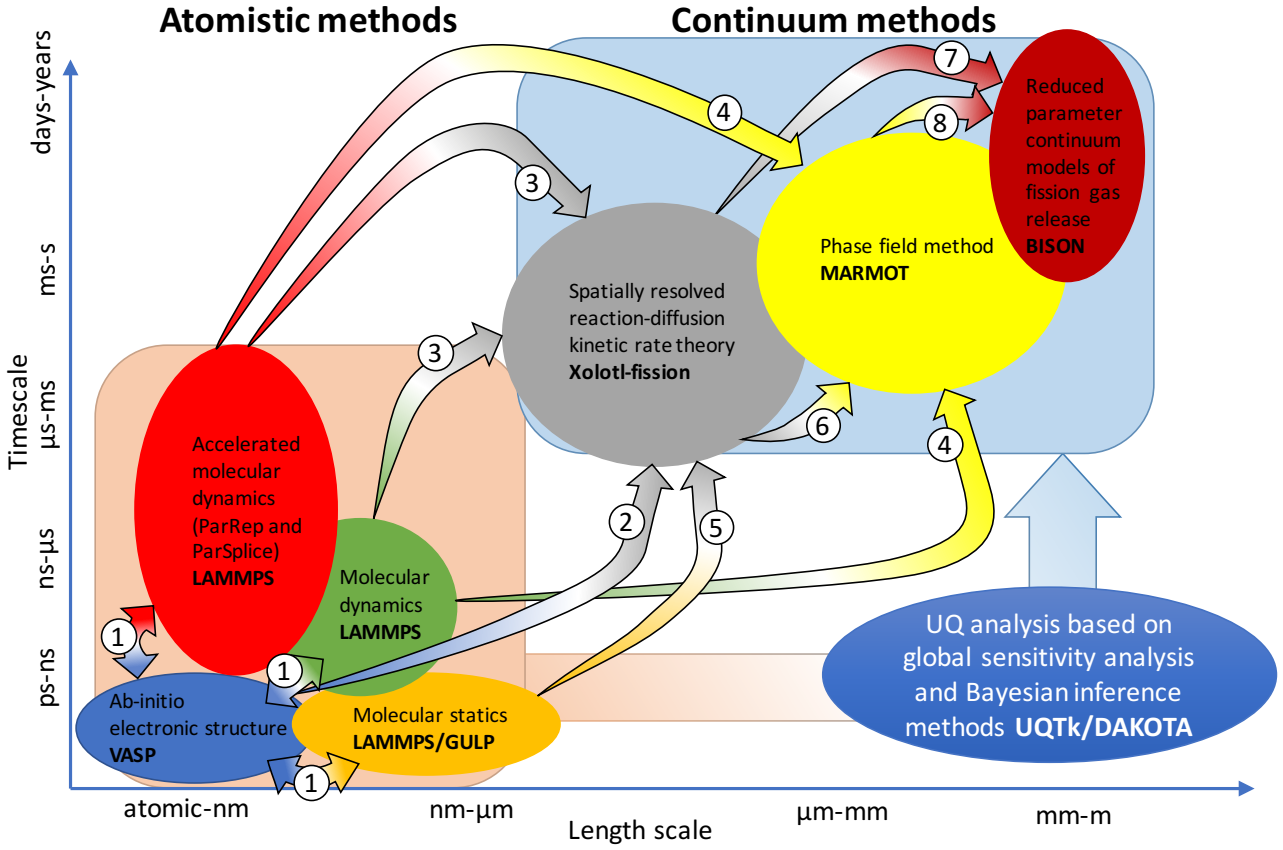
- Effective diffusion rate for accumulation at boundaries:

$$D' = Db' \Big/ \left(b' + g\right)$$

T. Matthews and D. Andersson (LANL)

Our vision of multiscale modeling and objectives

Although useful in steady-state simulations for known conditions, the current rate-theory model is not successful for transients, higher burnups, etc., because of the simplified geometric and physical representation.



Leverage accomplishments and tools developed by the DOE programs (NEAMS and SciDAC), extend to leadership-class computing and "multi-scale" UQ.

Utilize Cluster Dynamics spatially coupled to Phase Field Modeling informed by atomic scale simulations and uncertainty quantification to more accurately predict fission gas bubble populations & thereby and fission gas release.

Research activities organized in 3 thrusts

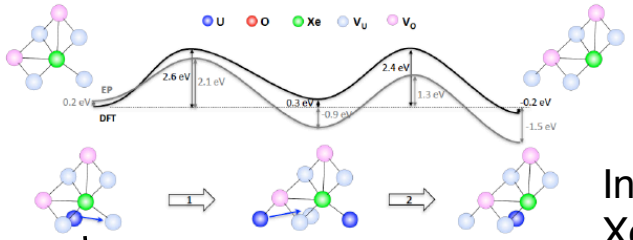
Computing, Computer Science and Applied Math (David Bernholdt)

- **Thrust 1 (David Andersson): DFT and long-time scale atomistic simulations to understand fission gas and defect behavior.**
  - Density Functional Theory (David Andersson)
  - Interatomic Potentials, AMD and MD Simulations Utilizing HPC (Danny Perez)
- **Thrust 2 (Brian Wirth): Spatially discretized cluster dynamics and MARMOT PFM simulations to understand fission gas bubble behavior.**
  - Xolotl-Fission Development and Coupling to MARMOT (Brian Wirth)
  - MARMOT Simulations of Inter-Granular Bubble Evolution (Mike Tonks)
  - Reduced order Model in BISON (Giovanni Pastore)
- **Thrust 3 (Habib Najm): Uncertainty quantification and experimental validation.**
  - UQ Methods (Habib Najm)
  - Experimental Validation (Giovanni Pastore)

Point defect and Xe diffusion from DFT and FECD

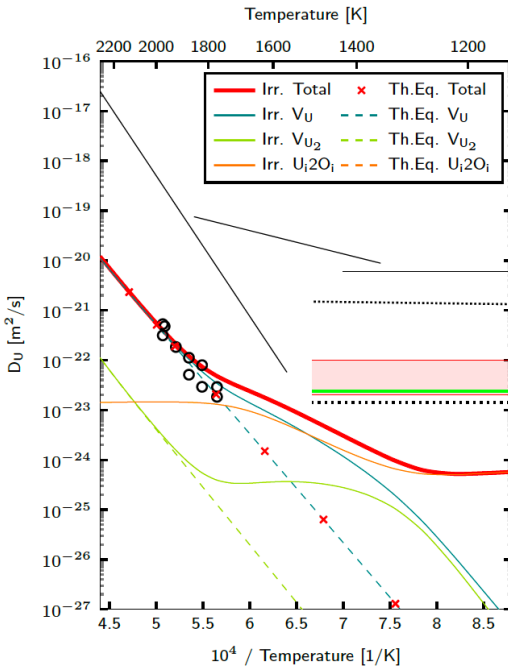
- DFT calculations to develop and parametrize a cluster dynamics model (FECD) for point defects and defect clustering in UO_2 to describe U and Xe diffusion under intrinsic and irradiation conditions[1,2,3].
- Predicts effective diffusion rates in UO_2, which is provided as input to MARMOT and Xolotl for simulation of intra- and inter-granular fission gas evolution.

The $4V_U$:$3V_O$ cluster dominates for D_2

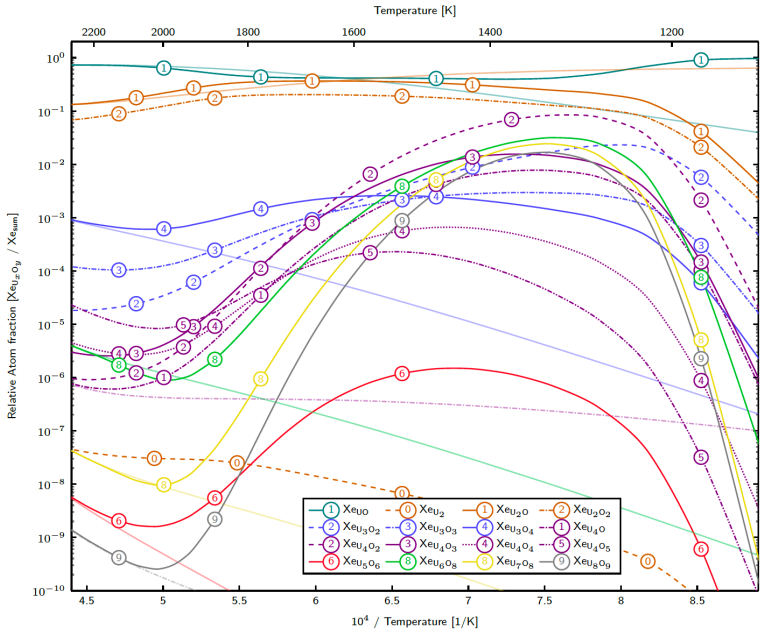


Input to Xolotl-MARMOT

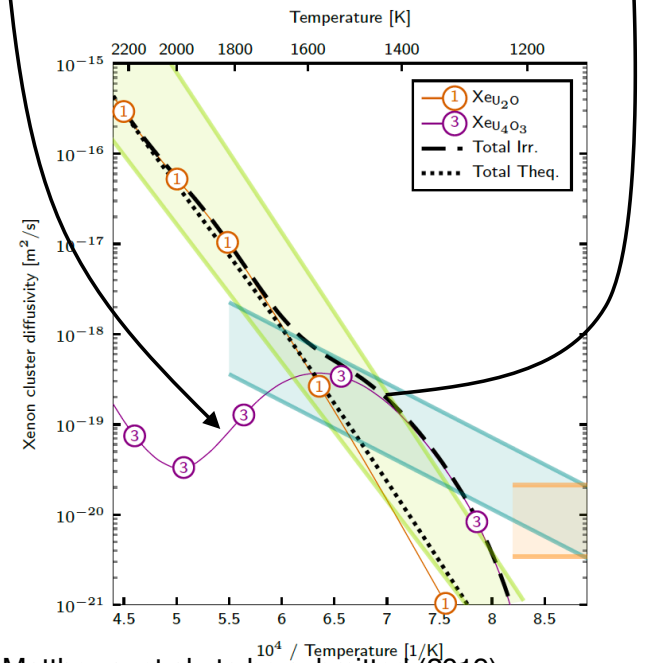
Uranium diffusion



Xe defect concentrations



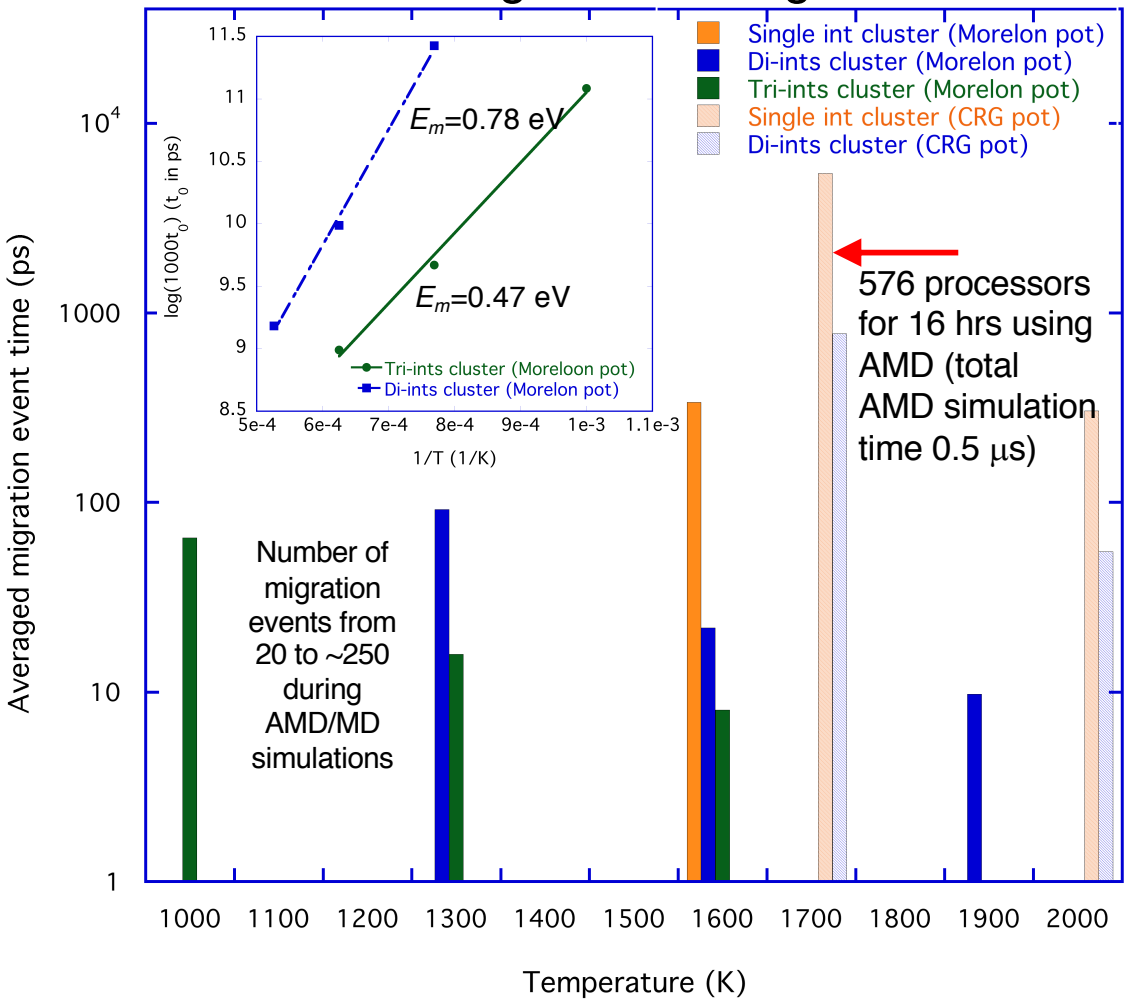
Xe diffusion



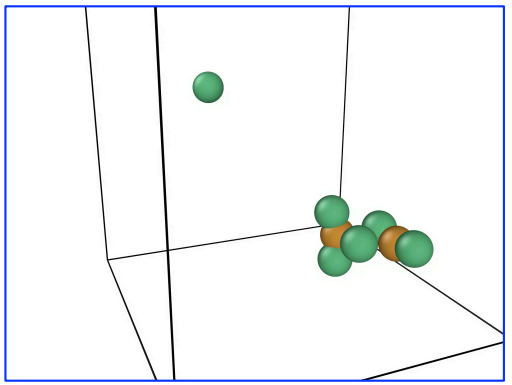
1. R. Perriot, et al., J. Nucl. Mater. **520**, 96-109 (2019). 2. C. Matthews, et al., under review (2019). 3. C. Matthews, et al., to be submitted (2019).

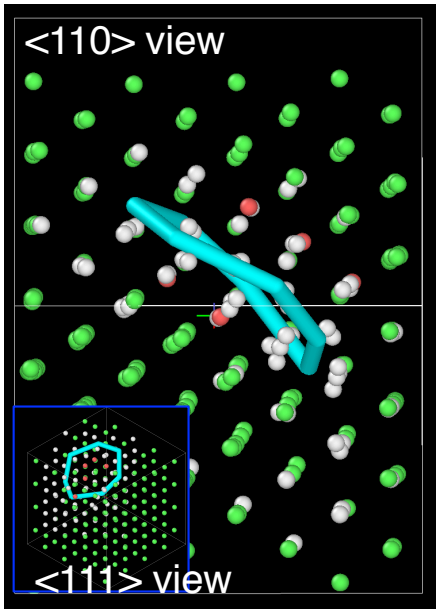
Interstitial cluster aggregates in UO_2 from MD/AMD

- Fast diffusion of U_i2O_i, formed under irradiation, by an interstitialcy mechanism.
- Accelerated molecular dynamics (AMD) and MD simulations using two empirical potentials to understand the cluster migration dynamics vs. cluster size.
- Even faster migration of larger interstitial clusters.



Tri-interstitial (anti-Schottky) cluster migrating towards single interstitial at 1000K





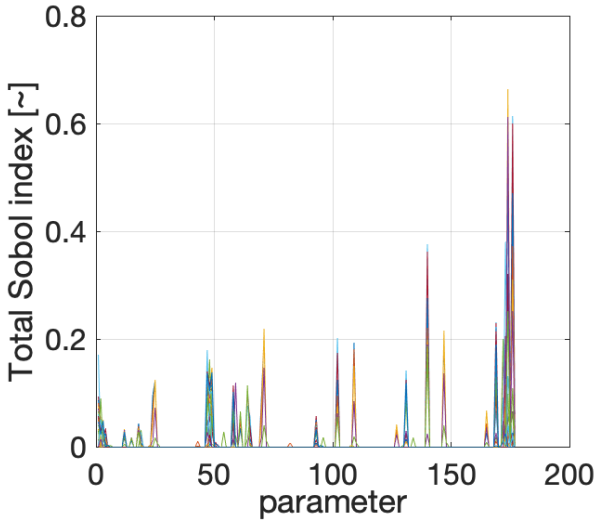
<111>/3 Frank loop forms via 10-interstitial cluster

Sensitivity and UQ analysis of FECD

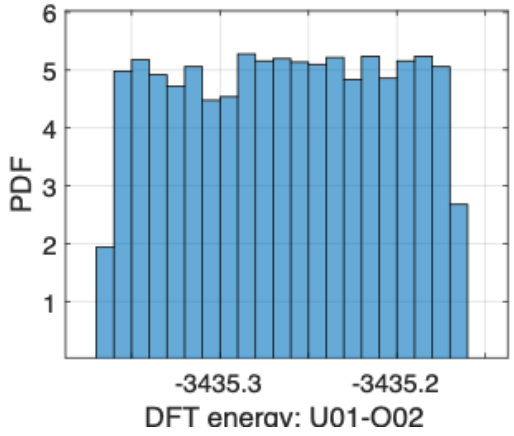
- The FECD cluster dynamics model is complex with inherent uncertainties for the parameter set and the current predictions involve by hand “calibration” to known experimental data.
- Use formal sensitivity and UQ analysis to understand the parameter influence as well as formalize “calibration” to a limited set of experiments.
- Ground work for full multi-scale calibration of MARMOT-Xolotl simulation of fission gas evolution informing reduced order engineering scale models.
- First perform sensitivity analysis to identify influential FECD input parameters, using uniform uncertainty specifications from expert opinion.
- Use a global sensitivity approach – accounting for variation of all parameters at all conditions (not just local perturbations).
- Multiple strategies for estimating global sensitivity indices, decide to use a Polynomial Chaos Expansion surrogate approach that assumes a degree of smoothness in the FECD mapping across input space.

Sensitivity indices of FECD extracted from PCE

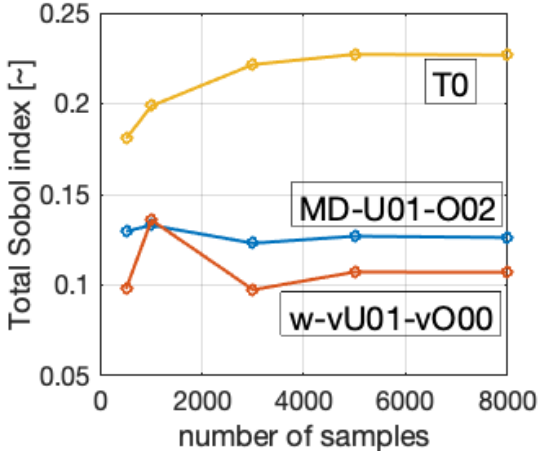
- Sensitivity (Sobol) indices extracted from coefficients of a PCE fit to FECD samples.
- PCE smoothness approximation - should require fewer FECD runs for sensitivity index convergence.
- PCE coefficients approximated using least-squares regression with random samples – can ignore sample points from failed runs.
- Expect many non-influential input parameters, so use a sparse regularization approach to efficiently find zero valued sensitivities (requiring fewer FECD runs).



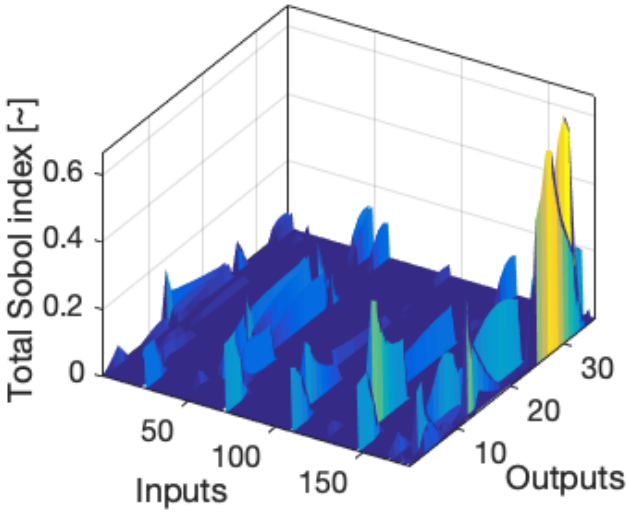
sensitivity indices



histogram of samples from uniform PDF on FECD parameter



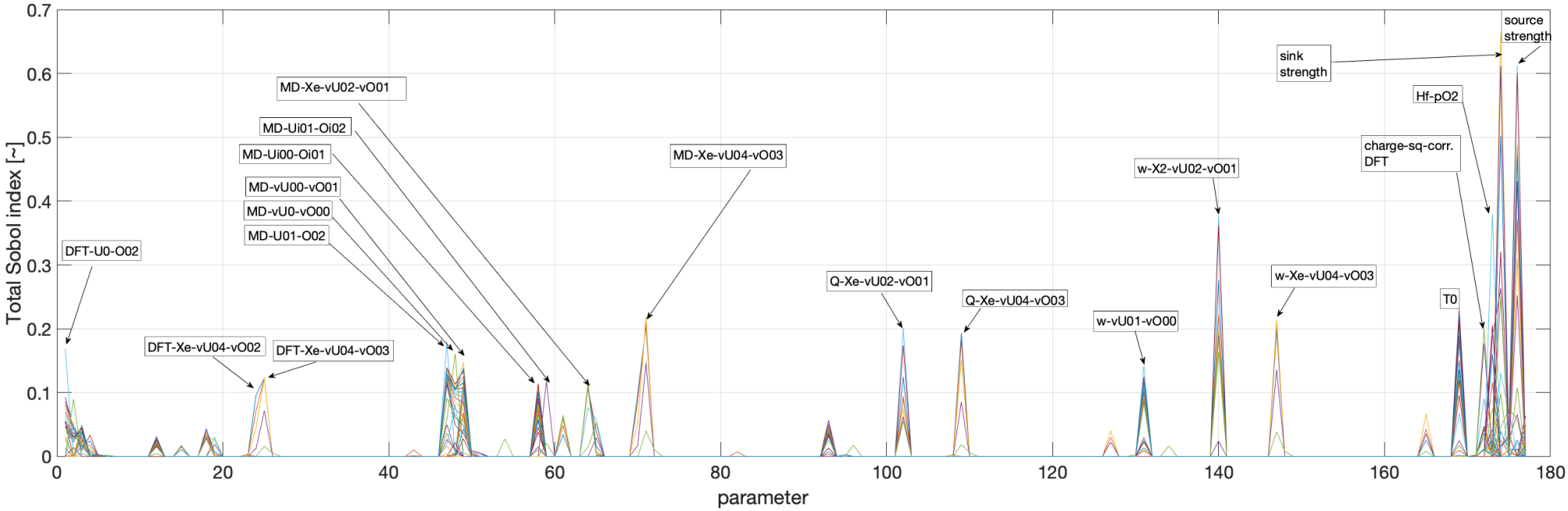
convergence of sensitivity index calculation for number of FECD runs



2D sensitivity index map across all quantities of interest

20 dominating parameters in FECD

- Identified on the order of 20 parameters (out of original 177) that dominate the model output variance across all diffusivities at all temperatures (36 quantities of interest).



- Can now construct a surrogate model taking only these 20 parameters (or fewer) into account.

UQ plans, targets and work in progress

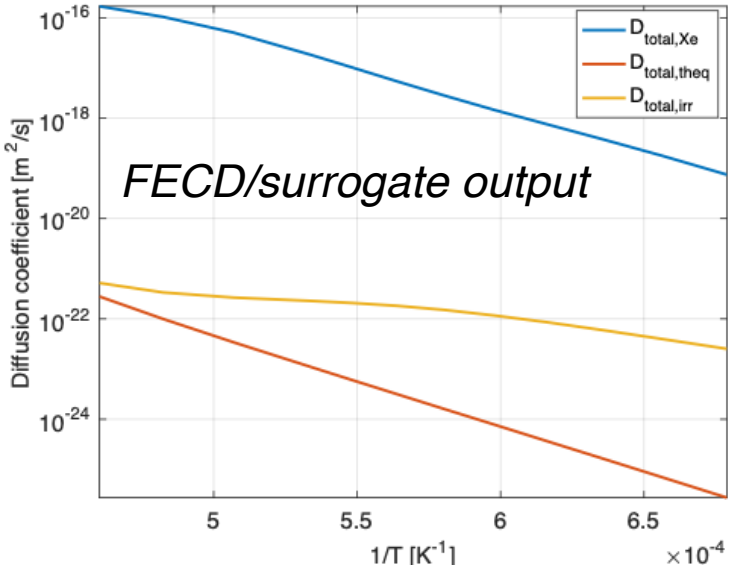
- The sensitivity analysis procedure involved building a surrogate model for the FECD computed diffusivities as a function of uncertain inputs (λ) at multiple temperatures using a polynomial expansion:

$$f(\lambda) \approx g_c(\lambda) = \sum_{k=0}^{K-1} c_k \Psi_k(\xi)$$

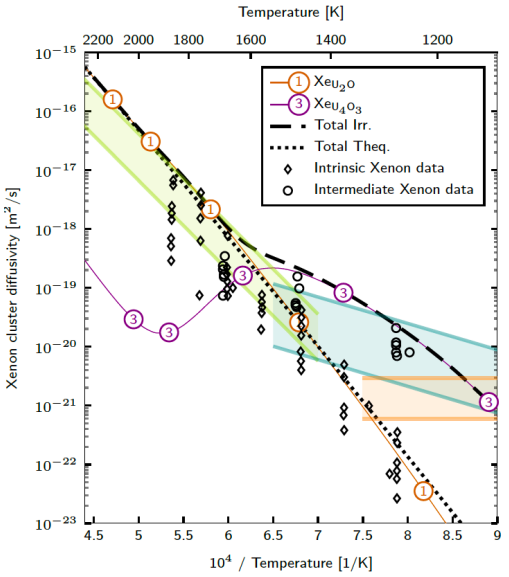
- Can use this polynomial to cheaply compute FECD outputs for specific inputs. Enables the solution of an inverse problem to calibrate particular parameters (β) against experimental data (z) using Bayesian inference:

$$p_{posterior}(\beta|z) = \frac{p_{likelihood}(z|\beta)p_{prior}(\beta)}{p(z)}$$

- Ultimately FECD predictions will be used to construct surrogates for Xolotl/MARMOT simulations in concert with other uncertain Xolotl/MARMOT input parameters with associated sensitivity analysis for down-selection.



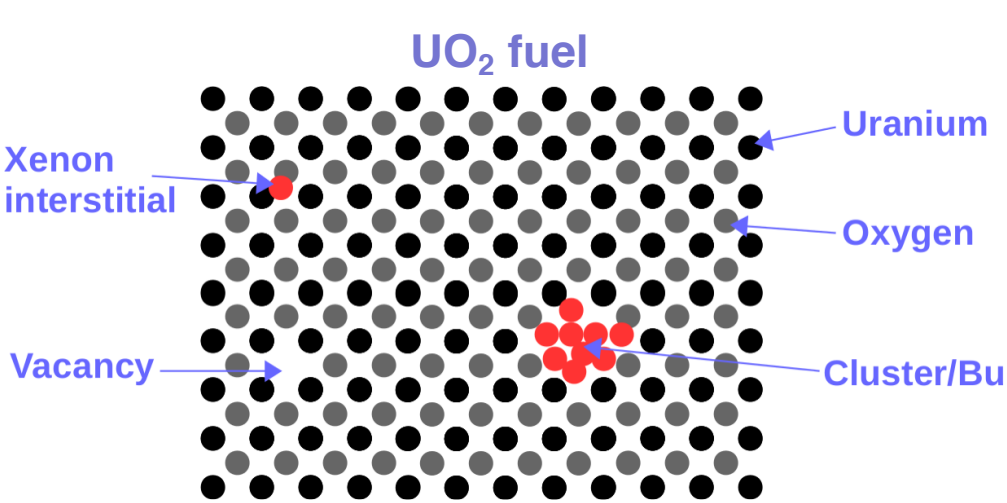
Experimental data

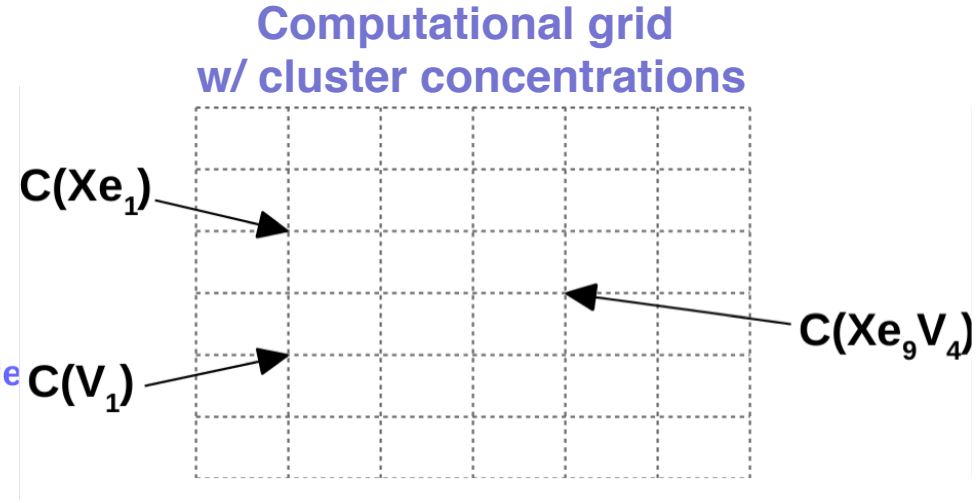


Xolotl, reaction-diffusion-advection cluster dynamics

- Xolotl (sho-lo-till) is the Aztec god of lightning and death
  - Started for plasma-surface interaction modeling for PSI fusion SciDAC (2012-2017)
- Spatially-resolved, time evolution of clusters of atoms, vacancies, interstitials within material based on kinetics
  - Including reaction, diffusion, advection, etc.
  - Material represented with a rectangular spatial grid (variable)
  - 0d, 1d, 2d, 3d models switchable at run time
- Bubble formation and evolution is major scientific focus (but not exclusive)

$$\delta_t \bar{C} = \phi \cdot \rho + D\nabla^2 \bar{C} - \nabla \bar{\nu} C - \bar{Q}(\bar{C})$$





Xolotl is available at https://github.com/ORNL-Fusion/xolotl/

Bubble re-solution model in Xolotl

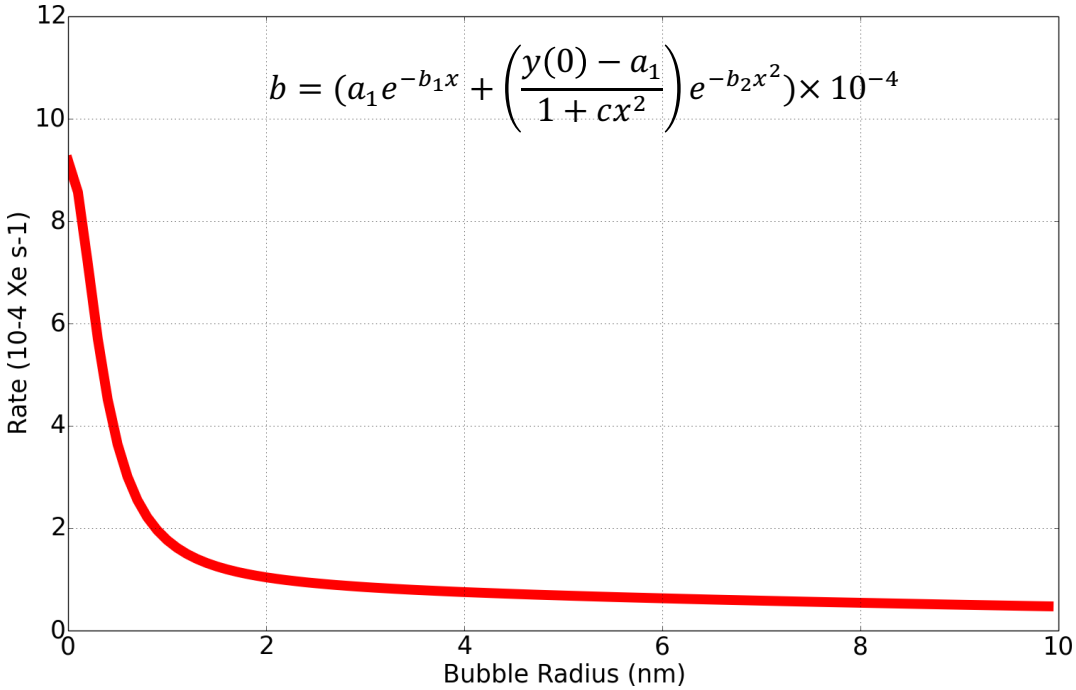
- The re-solution rate is used to model the reaction with a rate of k^-:

$$Xe_i \rightarrow Xe_{i-1} + Xe$$

$$k^- = b \times 10^8 \times 4 \times Xe_{rate}$$

- Initial implementation of re-solution slowed Xolotl by ~2X

Resolution rate from Xe bubbles from MD[1]

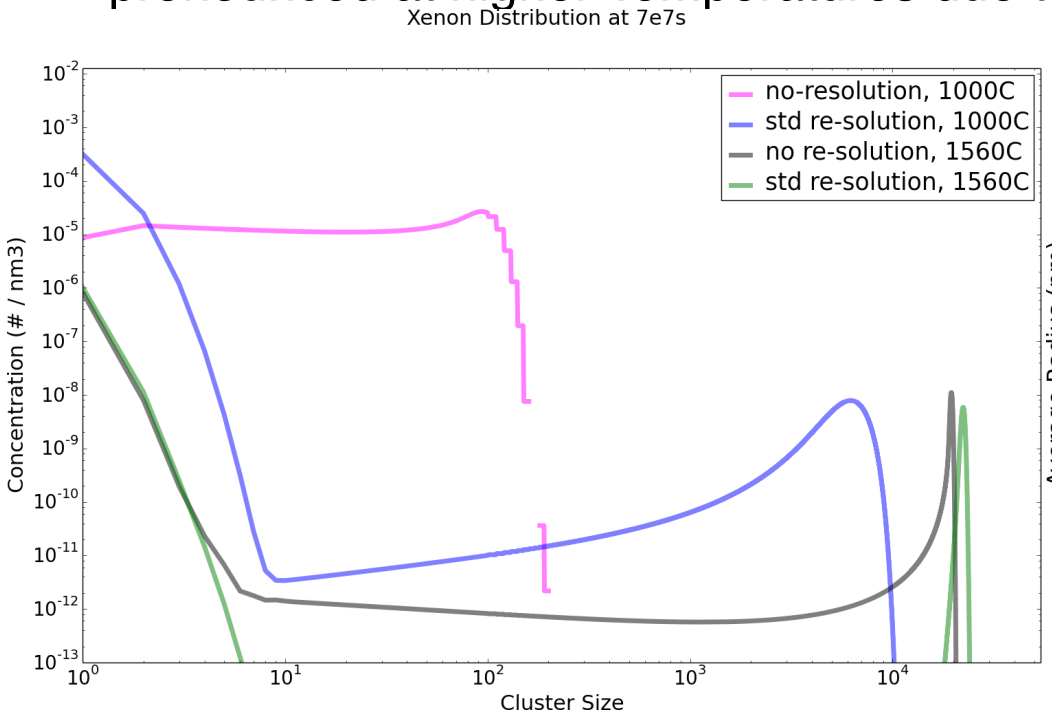


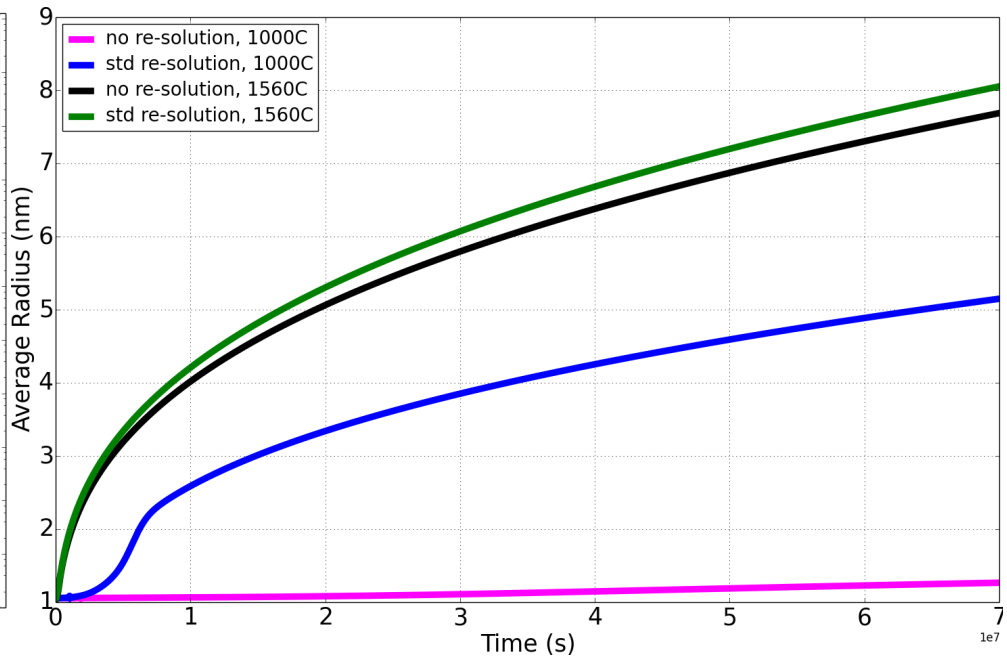
- Used the built-in performance infrastructure (developed by P. Roth) to identify that majority of the time difference was spent in computing and setting the Jacobian elements.
- Switched to setting the Jacobian elements in PETSc in batches instead of individually.
- The difference in timing is now only 1.2 times slower.

1. W. Setyawan, et al., J. App. Phys. **124**, 075107 (2018).

Impact of gas bubble re-solution in Xolotl

- Implementation of the homogenous re-solution model leads to significant modification to the Xenon bubble size distribution (for nominal fission rate density of 10^{19} fissions m^{-3} s^{-1}) at 1000° C, and to a lesser extent at 1560° C at a time of $7x10^{7}$ seconds (~3% burnup).
- Size distribution hints at beginning stages of a bi-modal size distribution, especially at lower Temperatures but also results in a larger average size (less pronounced at higher Temperatures due to faster Xe diffusion).





- Continuing efforts to assess key sensitivities in model & benchmark to experimental data for validation.

MARMOT meso-scale fuel performance tool

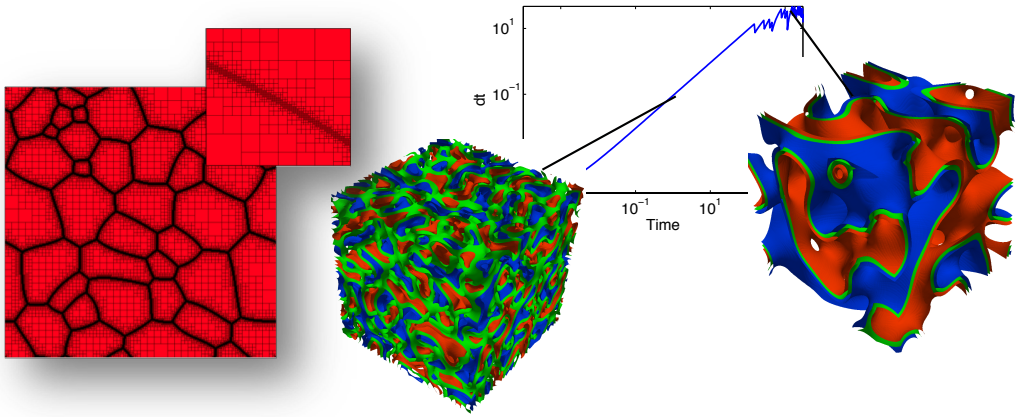
- **Predicts the coevolution of microstructure and properties in nuclear materials.**

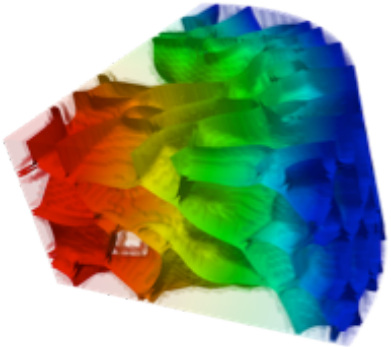
Technique: Phase field coupled with large deformation solid mechanics and heat conduction solved with implicit finite elements using INL's MOOSE framework

MOOSE

MARMOT:

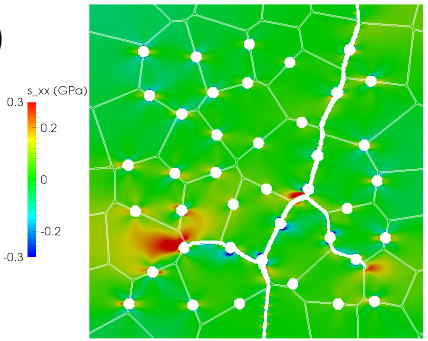
- Uses FEM with implicit time integration
- Built on the **LibMesh** FEM library
- 1D, 2D, or 3D without recompile
- System is solved using Newton or JFNK (GMRES) via **PETSc**
- Employs mesh and time step adaptivity





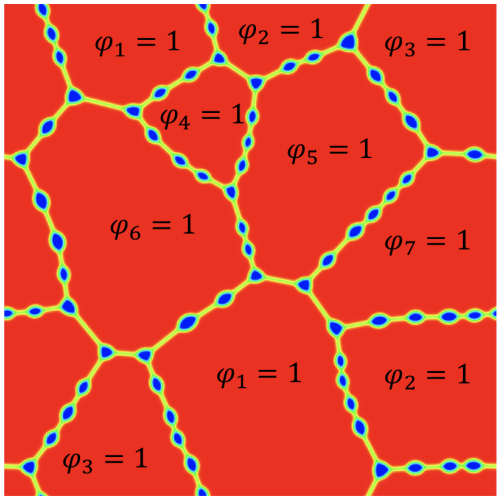
Physical models include:

- UO_2 (sintering, grain growth, fission gas, fracture)
- U-Zr (species transport, phase change, swelling)
- U-Si (grain growth, fission gas, swelling)
- Zircaloy cladding (Hydride formation)
- FeCrAl cladding (Creep, Swelling)

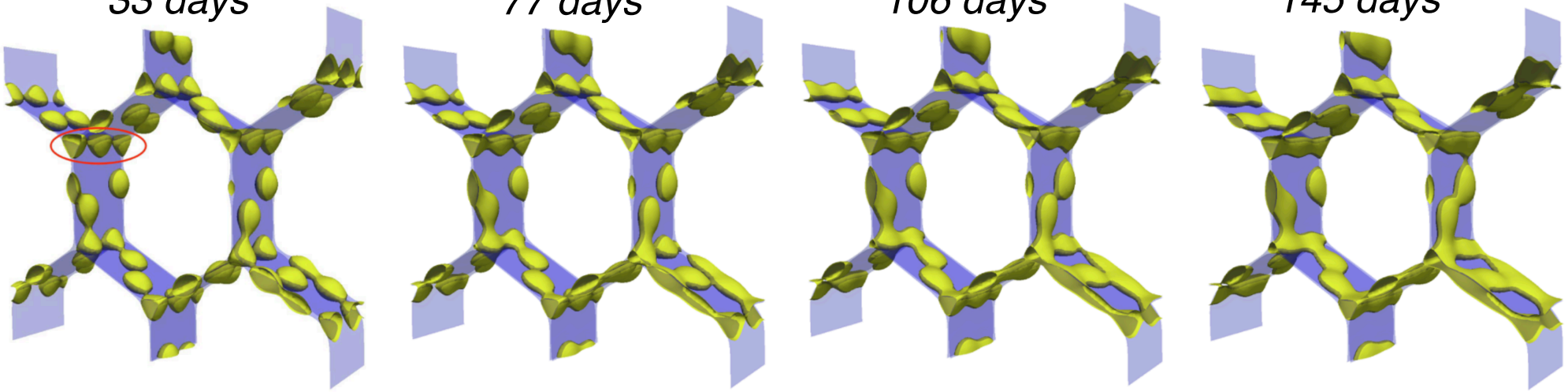


The MARMOT fission gas model

- The model predicts the growth and coalescence of fission gas bubbles within grains and on grain boundaries to better understand fission gas release.
- MARMOT fission gas model predicts:
  - Transport of gas atoms and U vacancies (2 DOF).
  - Void growth and coalescence (1 DOF).
  - Grain boundary migration (1 – 20 DOF).
- The model uses 4 – 22 DOF per node.
- From Aagesen et al. Comp. Mat. Sci. 161 (2019): 35-45.

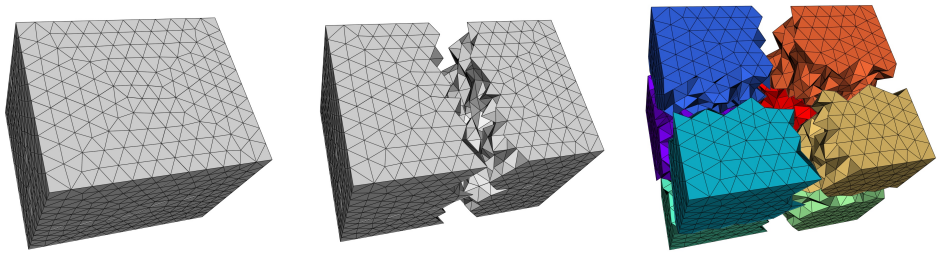


33 days *77 days* *106 days* *145 days*

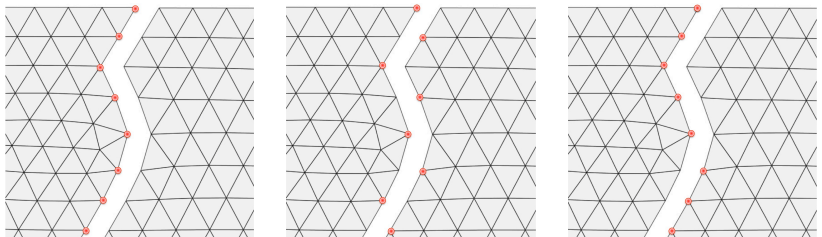


MARMOT performance and scalability

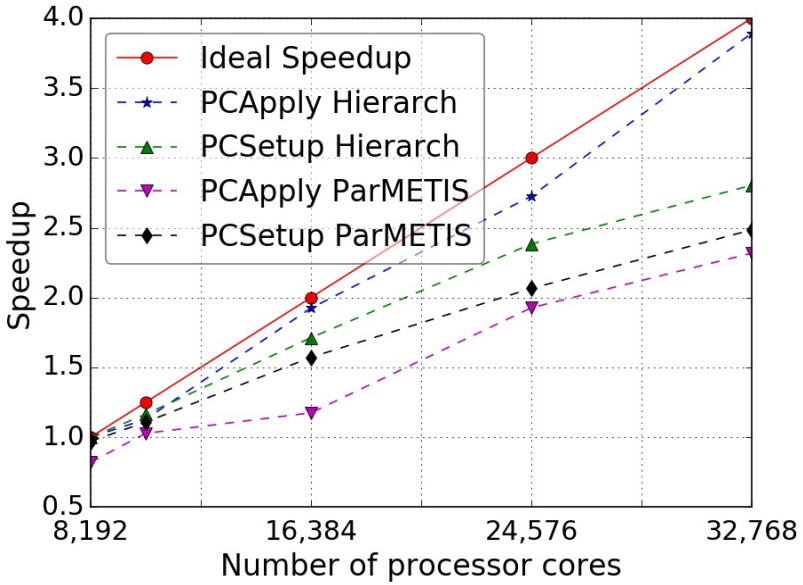
- MARMOT scalability using > 8000 processors has been improved through the development of a hierarchical partitioning algorithm in PETSc and a novel node assignment algorithm in libMesh.

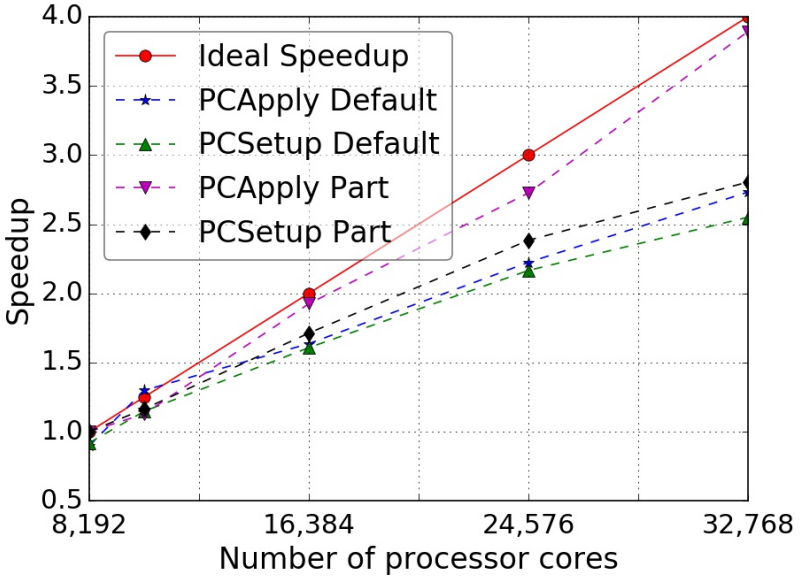


New PETSc hierarchical partitioning algorithm partitions graph into subgraphs that are then partitioned into smaller subgraphs .



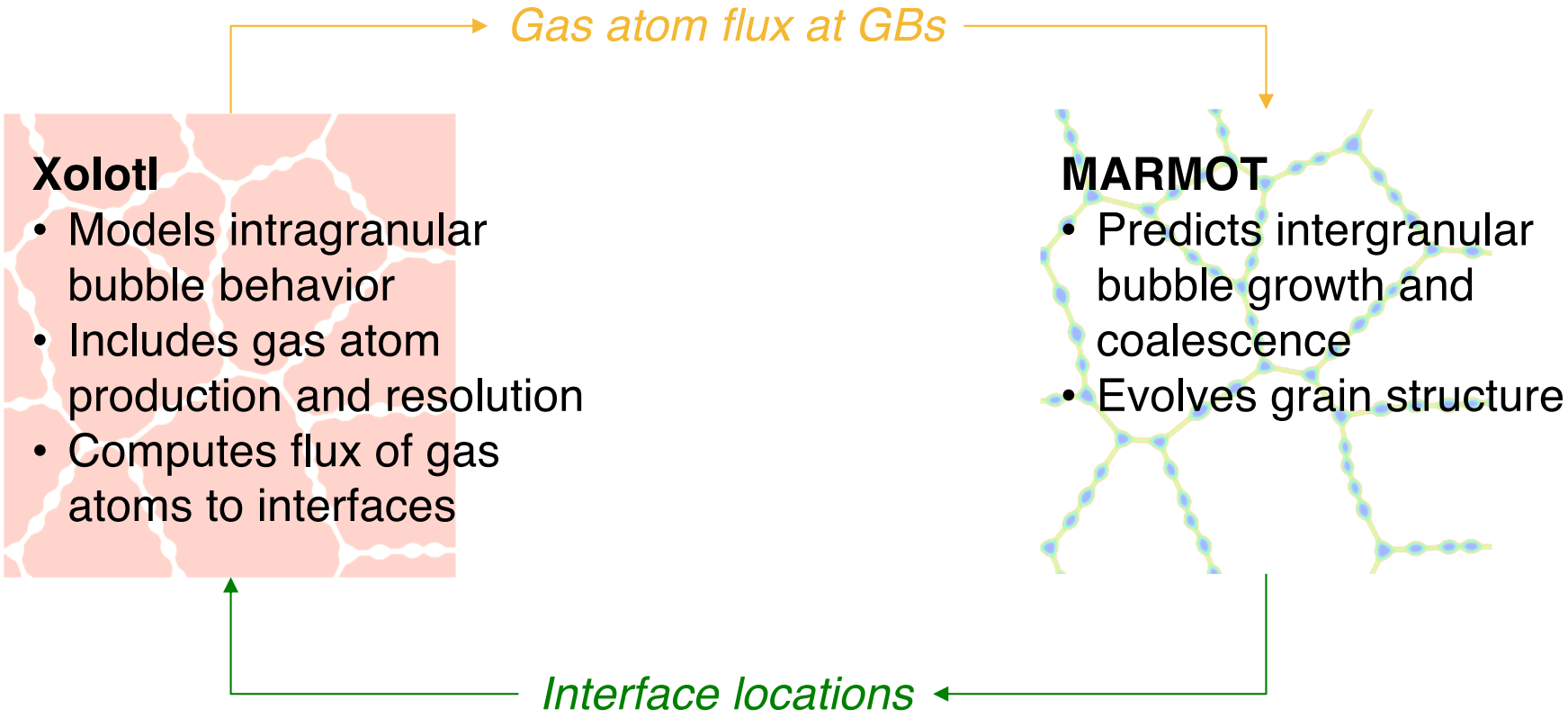
New libMesh node assignment algorithm applies partitioner to neighboring MPI processes and assigns one submesh to neighboring MPI ranks.





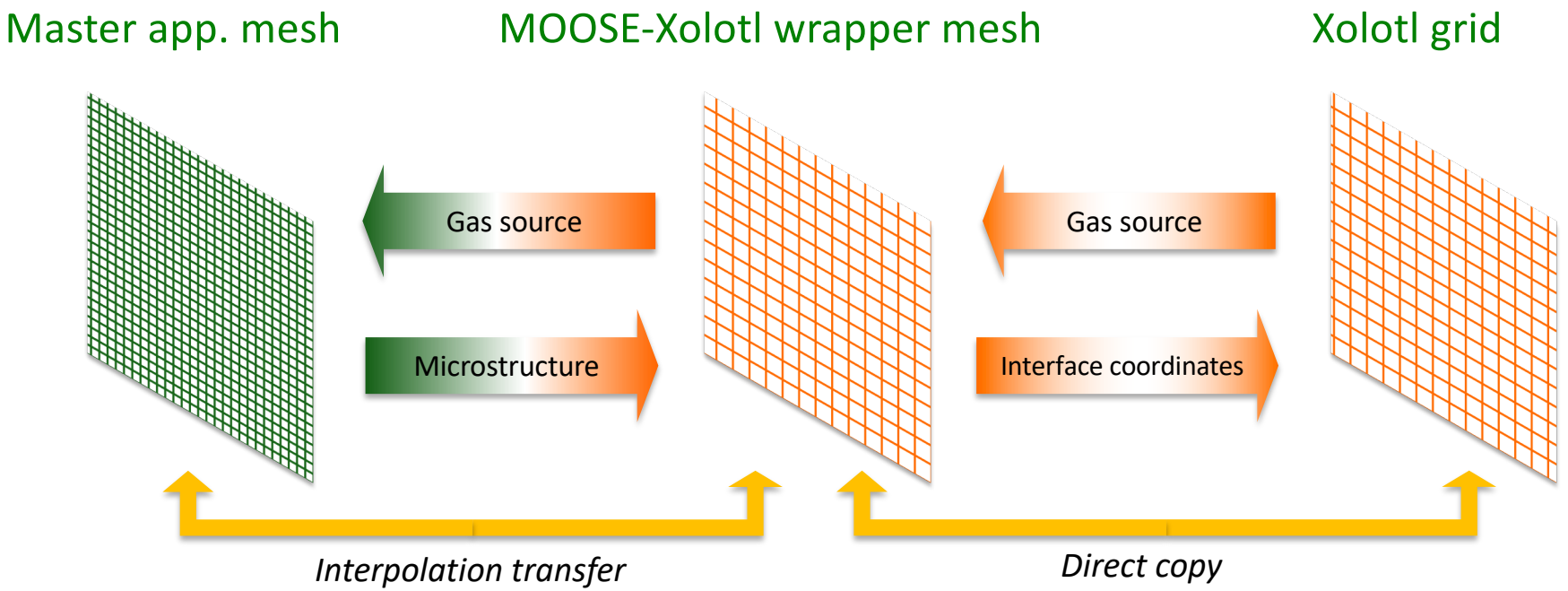
Xolotl-MARMOT coupling: Approach

- The coupled code uses
  - cluster dynamics in Xolotl to consider fission gas transport bubble behavior within grains.
  - Phase field method in MARMOT to consider intergranular fission gas bubbles.



Xolotl-MARMOT coupling: Mesh interpolation

- The coupling is managed using the MOOSE ‘MultiApps’ and ‘Transfers’ systems.



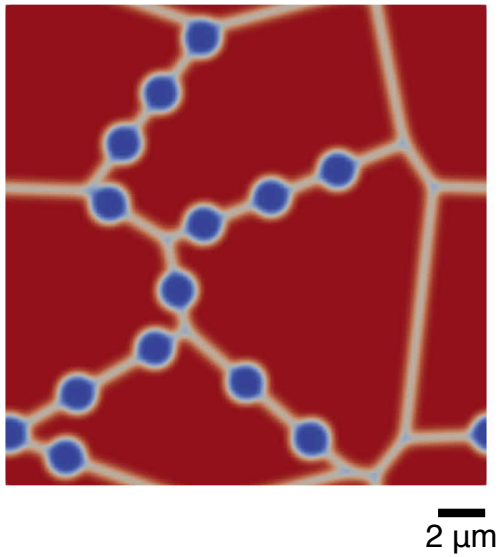
- Data transfer between nonidentical mesh.
- Mesh adaptivity is available in MARMOT.

- Positions of every node should be identical.
- Parallelism is identical and data passing is done locally on each node.

Xolotl-MARMOT coupling: Initial results

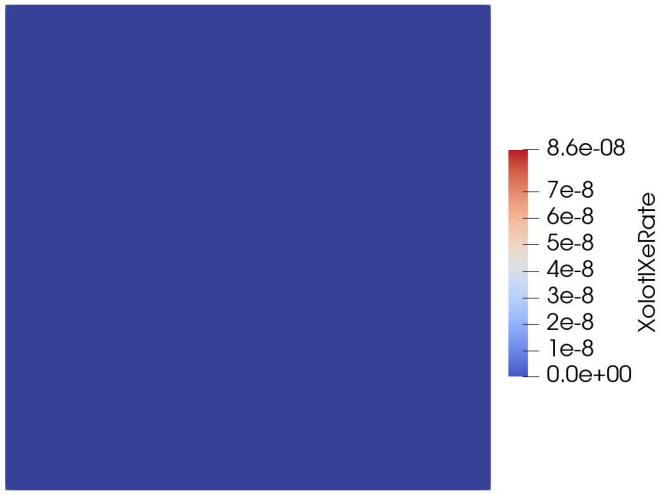
- We have coupled the two codes to model fission gas behavior in 2D polycrystalline UO_2.

Time: 0.000000e+00 s



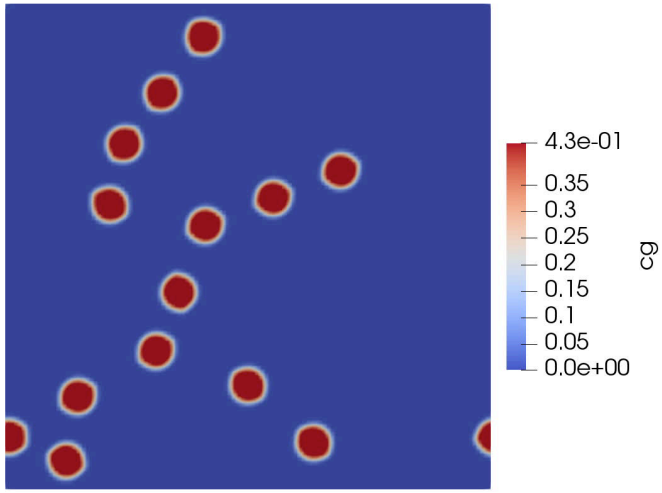
Microstructure

Red: fuel
Blue: bubble



Passed from Xolotl

Xe generation rate

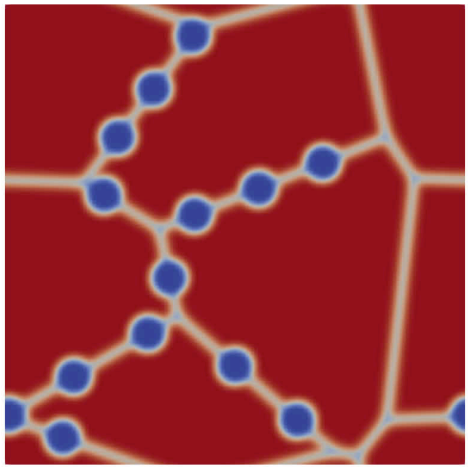


Xe concentration

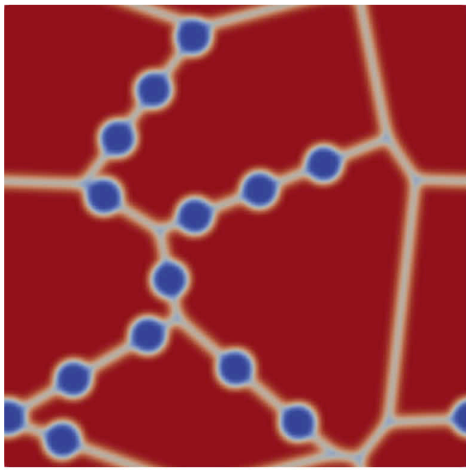
Xolotl-MARMOT coupling: Model comparisons

- Capturing more accurate physics in Xolotl directly changes MARMOT predictions

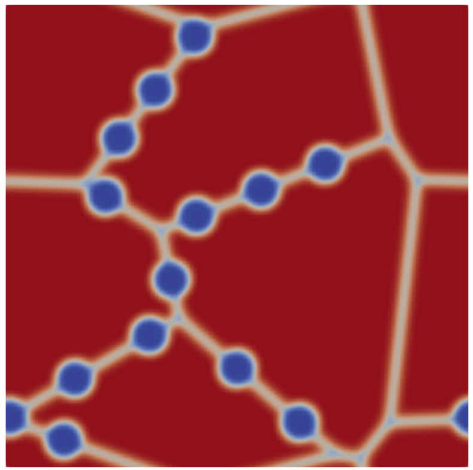
Time: 0.000000e+00 s



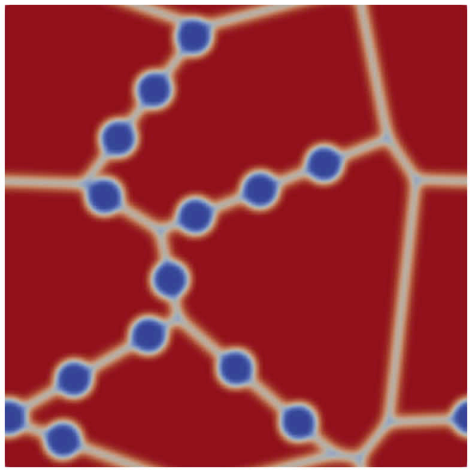
Only MARMOT



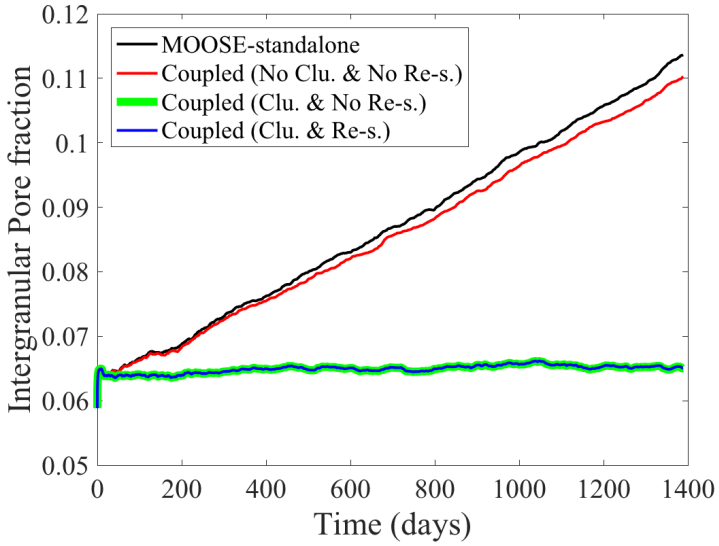
No Clustering & No Re-solution



Clustering & No Re-solution



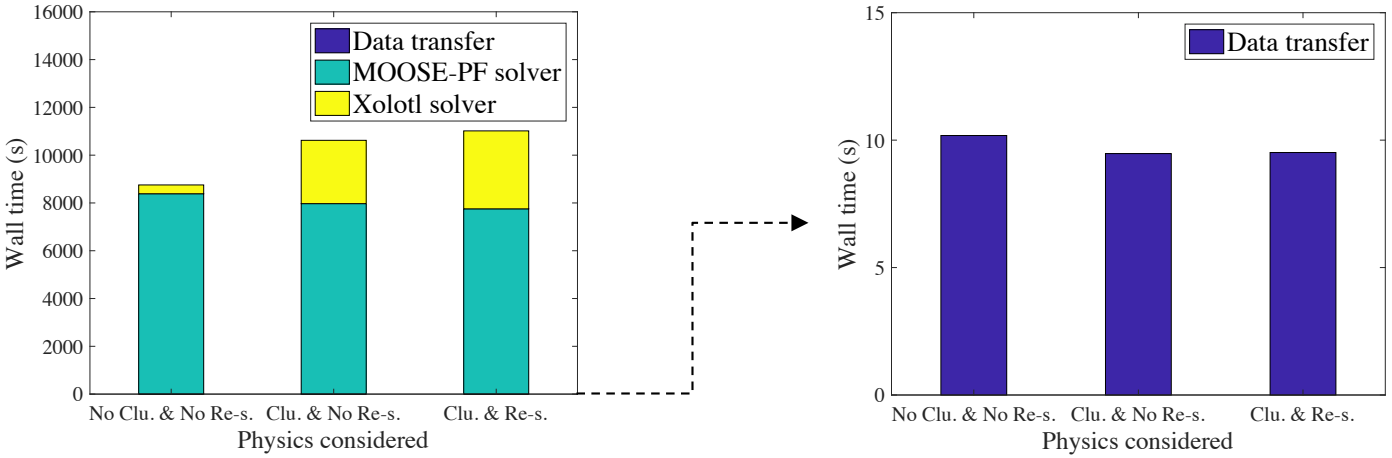
Clustering & Re-solution



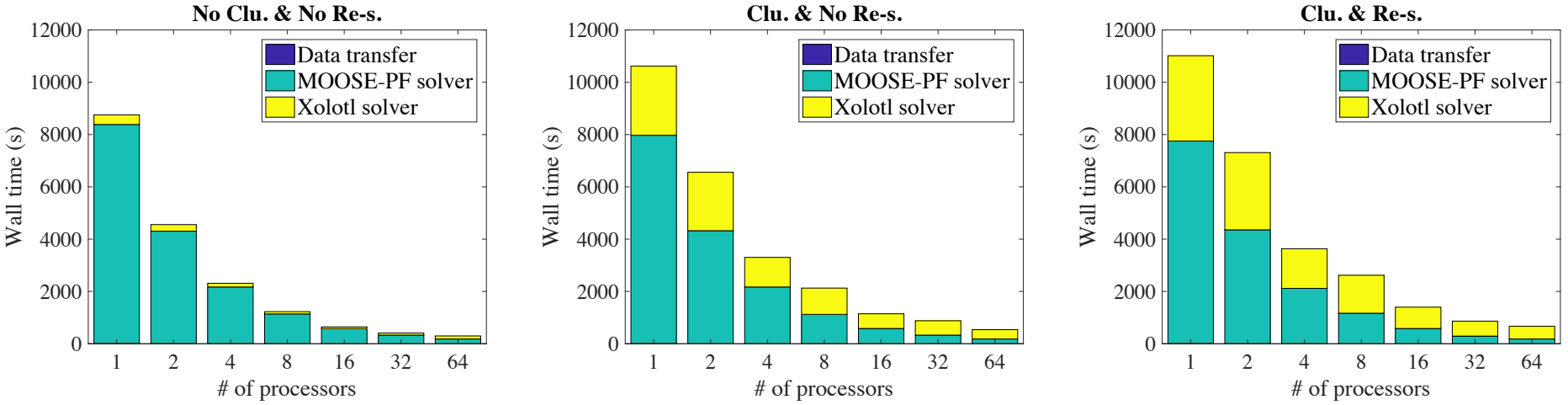
Xolotl-MARMOT coupling: Performance analysis

- Division of computation time between codes:
  - The data transfer between coupled codes costs negligible time (~1% or less).
  - Addition of physics (clustering & re-solution) changes overall computation time and division.

UF HyperGator acknowledged for computer resources.

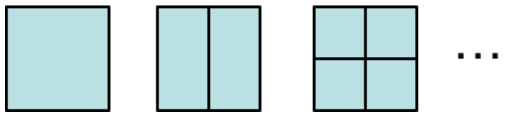


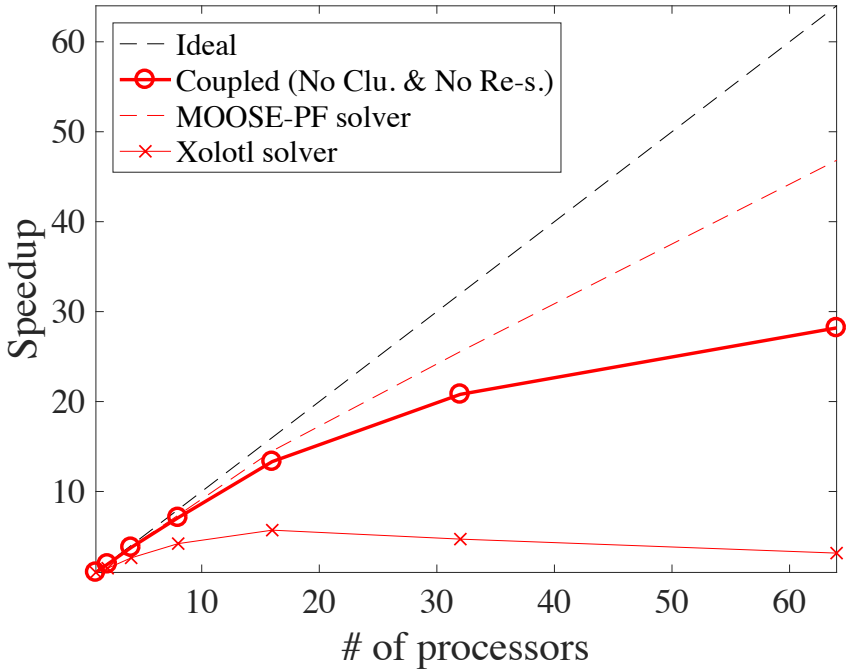
  - The # of processors is also a factor changing the division of computation time

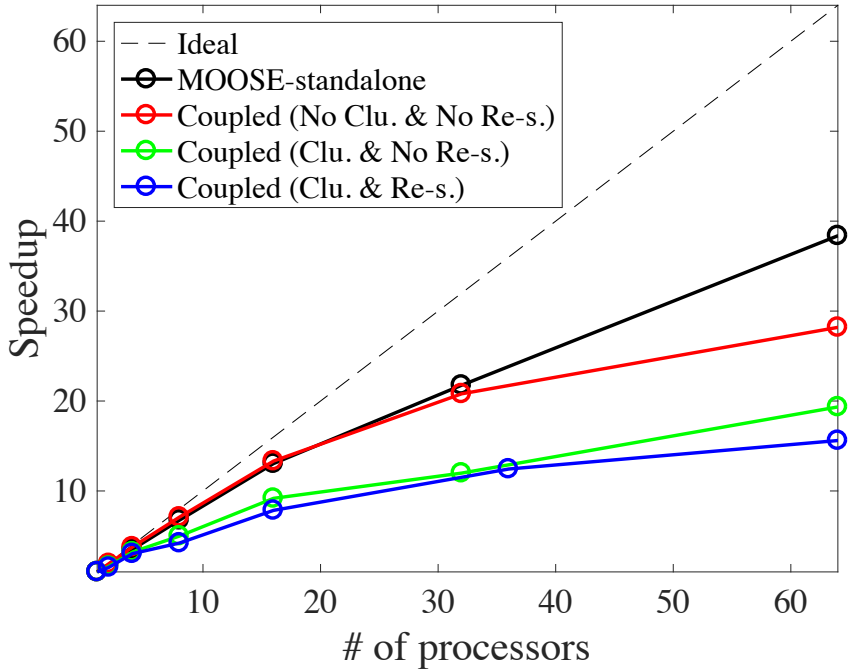


Xolotl-MARMOT coupling: Strong scaling

- Initial assessment of scalability of the coupled code was carried out.
- Strong scaling:
  - 2D, 125 x 125 elements with 126 x 126 nodes
  - # of DOF per node = 9*
  - Variable: # of processors



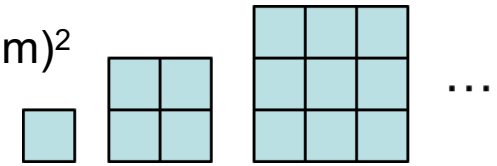


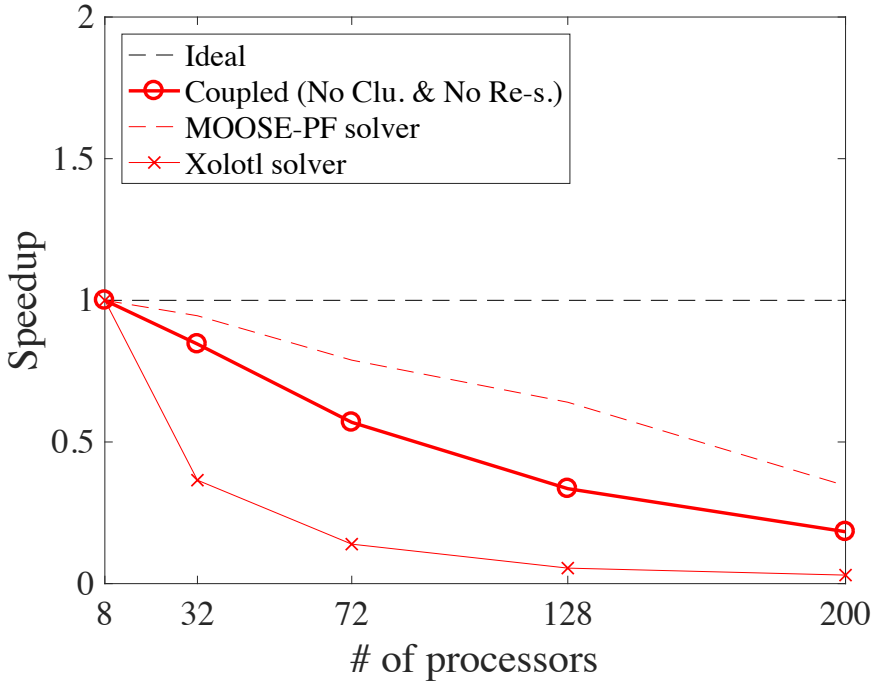


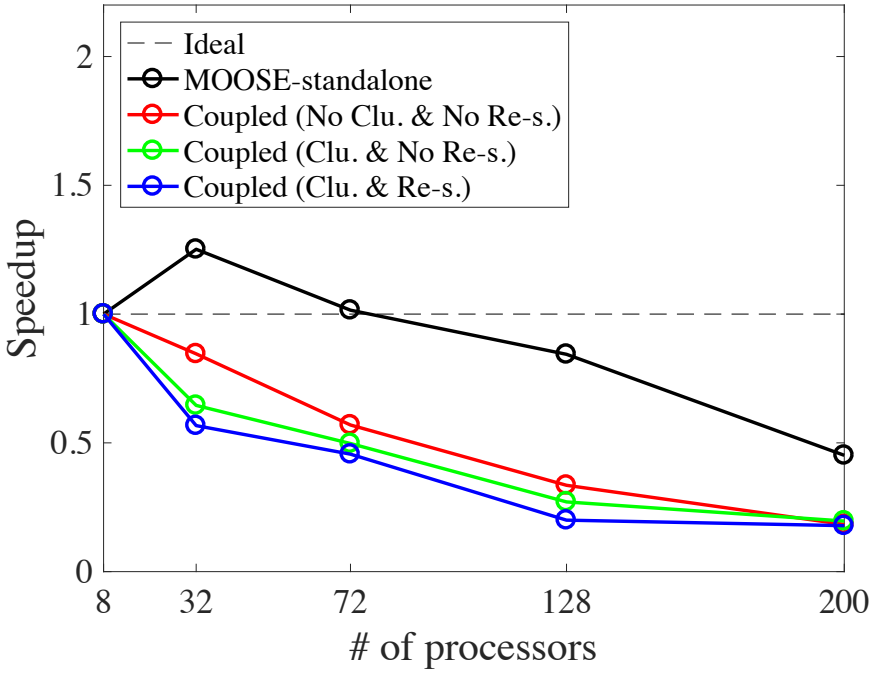
*If clustering is considered, # of DOF = 1008

Xolotl-MARMOT coupling: Weak scaling

- Weak scaling
  - # of nodes per area of simulation domain: 32.1 μm^{-2} (fixed)
  - Problem sizes: (9 $\mu m)^2$, (18 $\mu m)^2$, (27 $\mu m)^2$, (36 $\mu m)^2$, (45 $\mu m)^2$
  - # of processors: 8, 32, 72, 128, 200

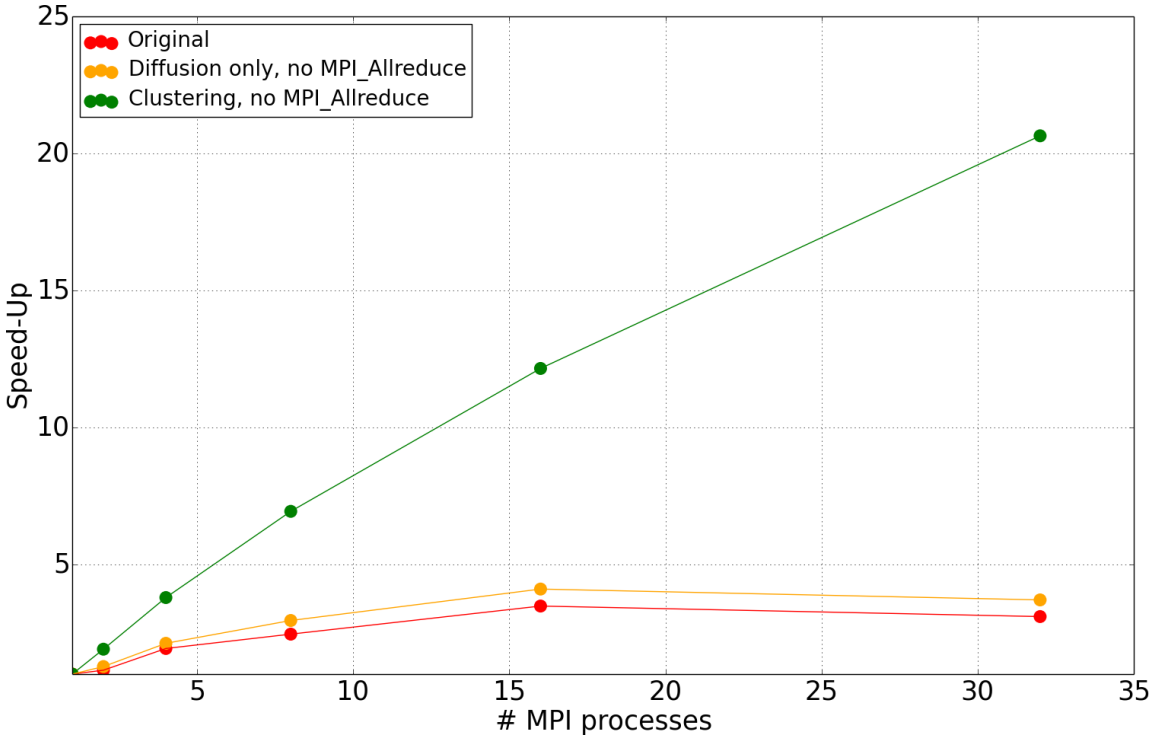






Xolotl strong scaling and improvements

- First strong scaling performed in 2D.
- Used hpctoolkit to identify a MPI_Allreduce() call that was taking most of the time.
- This call is needed in the fusion application Xolotl was originally developed for (the solver is common for all the applications).
- Added an option to skip this part of the solver when it is not needed.

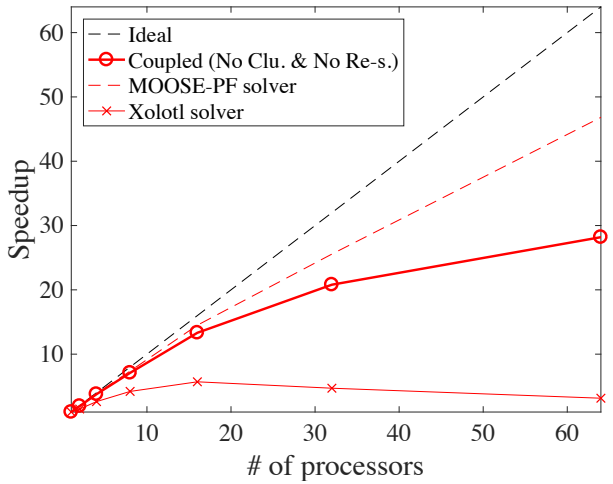


- After optimization, when only diffusion is happening (no clustering), hpctoolkit shows that what takes the most time is applying the pre-conditioner in PETSc.

Xolotl-MARMOT coupling: Future work

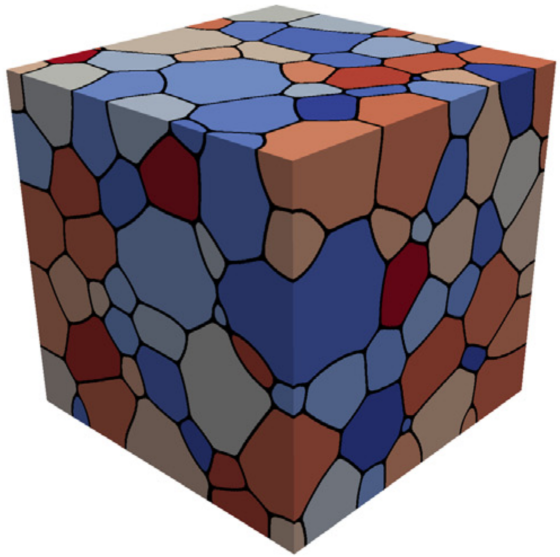
- Now that we have demonstrated the coupled code in 2D, we will make improvements to enable 3D simulations
- Future work:

We will improve the parallel scalability of Xolotl

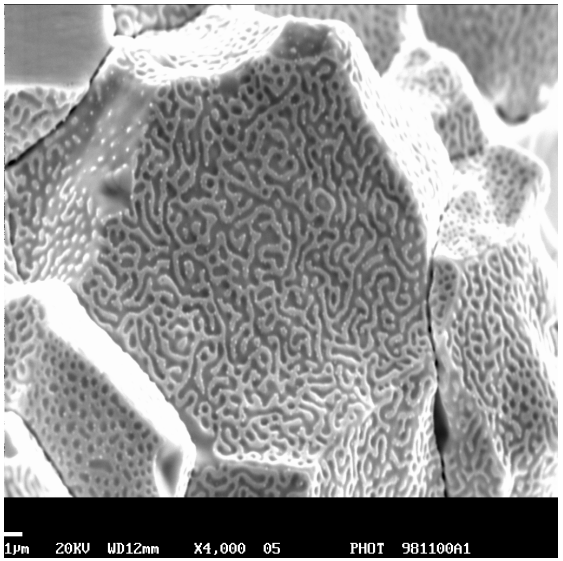


Progress already accomplished!

We will investigate the performance of the coupled model in 3D



We will validate the model by comparing to published data



Engineering model of intra-granular bubble evolution

$$\frac{\partial N_b}{\partial t} = \nu - \alpha_{\overline{n_b}} N_b$$

$$\frac{\partial m_b}{\partial t} = 2\nu + \beta_{\overline{n_b}} N_b - \alpha_{\overline{n_b}} m_b$$

$$\frac{\partial c_1}{\partial t} = yF + D\nabla^2 c_1 - 2\nu - \beta_{\overline{n_b}} N_b + \alpha_{\overline{n_b}} m_b - \beta' c_1 - \beta_{\overline{n_d}} N_d + \alpha_{\overline{n_d}} m_d$$

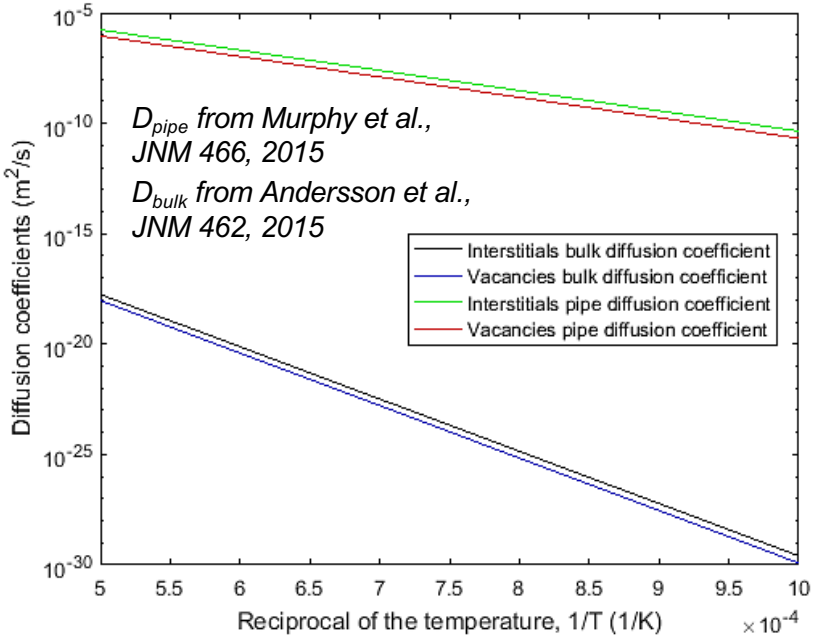
$$\frac{\partial N_d}{\partial t} = z\rho_d \delta(t - t_0) - \alpha_{\overline{n_d}} N_d$$

$$\frac{\partial m_d}{\partial t} = \beta' c_1 + \beta_{\overline{n_d}} N_d - \alpha_{\overline{n_d}} m_d$$

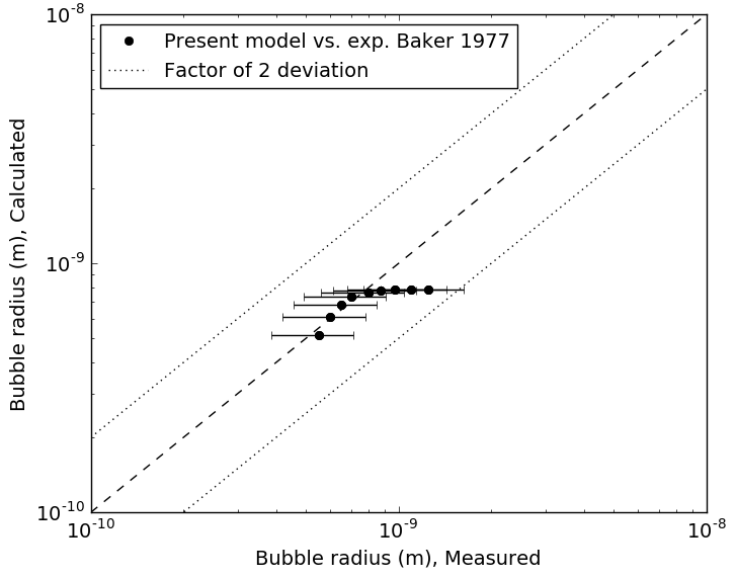
- Considers nanometric bubbles in the bulk (*b*), coarsened bubbles at dislocations (*d*, highlighted terms) and dissolved gas atoms (c_1).
- Bubble coarsening important for gaseous swelling during transients/high burnup.
- Vacancy inflow at coarsening bubbles through pipe diffusion along dislocations:

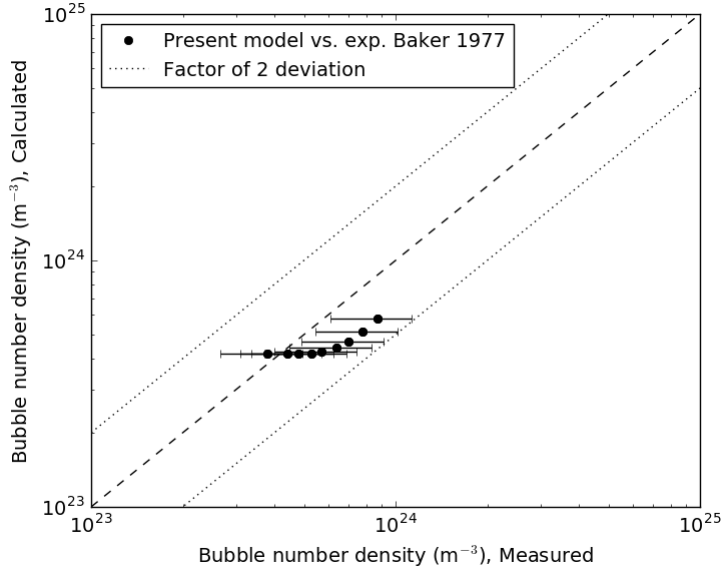
$$\frac{\partial \overline{n_v}}{\partial t} = \frac{2\pi D_{v,pipe} b}{kTs}\left(\frac{kT}{\Omega}\frac{\overline{n_d}}{\overline{n_v}} - p_{eq}\right)$$

- Multiscale coupling for diffusivities

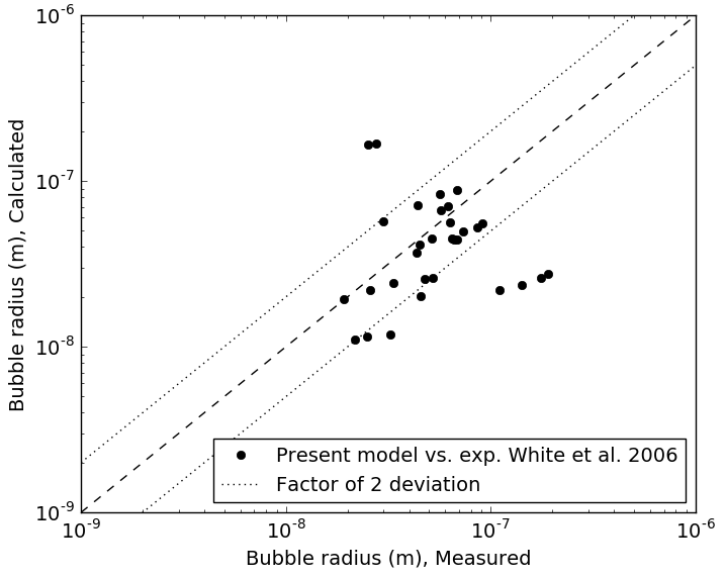


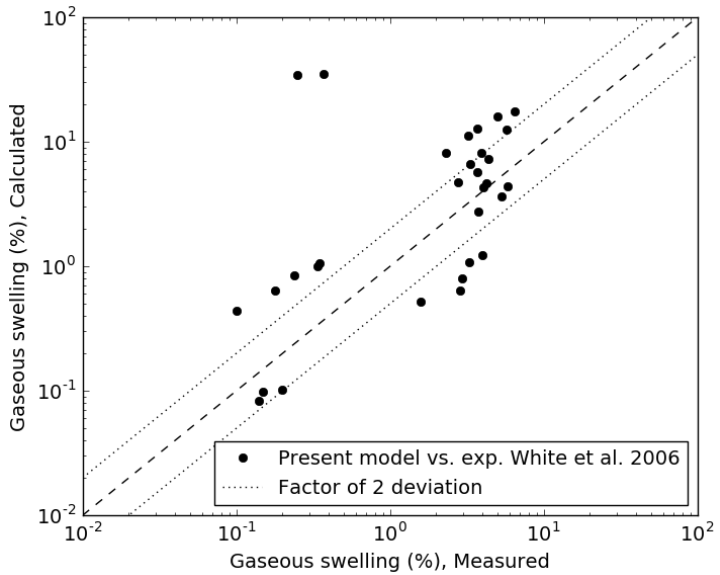
Experimental validation





Nanometric bubble size and number density in base-irradiated UO_2. Model vs. local data in Baker, JNM 66, 1977





Coarsened bubble size and swelling in ramp-tested UO_2. Model vs. local data in White et al., R&T/NG/EXT/REP/0206/02, 2006

Interaction with SciDAC institutes

- FastMath:
  - Xolotl solver-related issues and performance optimization.
  - UQ analysis of FECD and initial work on Xolotl.
- RAPIDS and FastMath:
  - Software engineering guidance for Xolotl-MARMOT coupled code, e.g.:
    - Packaging of Xolotl as a library (as opposed to executable) so its functions can be called by MOOSE framework.
    - Integration of two instances of PETSc in same program.
  - Performance analysis and optimization of Xolotl.

Summary and conclusions

- Progress highlighted in all 3 technical thrusts for the project “Simulation of fission gas in uranium oxide nuclear fuel”.
  - DFT and long-time scale atomistic simulations:
    - Atomic scale simulations based on DFT, MD and AMD identified mechanisms responsible for Xe diffusion and predicted response as function of temperature, chemistry and irradiation.
    - AMD simulations of uranium interstitial cluster formation and diffusion.
  - Spatially discretized cluster dynamics and MARMOT PFM simulations:
    - New fission gas resolution model added to Xolotl.
    - Performance optimization of both Xolotl and MARMOT to prepare for large-scale 3D coupled simulations.
    - Initial Xolotl-MARMOT coupling accomplished through the MOOSE multi-app system.
    - Application to physically accurate 2D problem.
    - Performance analysis identified areas for future optimization as we target large-scale 3D coupled simulations.
  - Developed engineering scale framework to be informed by the meso-scale models in future years and identified experimental validation targets.
  - Performed sensitivity analysis of bulk Xe diffusion predicted by the FECD code using PCE methodology.