# *Simulation of fission gas in uranium oxide nuclear fuel*

**David Andersson* on behalf of**

| **Institution** | **Principal Investigator** | **Additional personnel** |
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| U. Tennessee | Brian Wirth | Abdullah Al-Fadhili, Sophie Blondel |

In partnership with:

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

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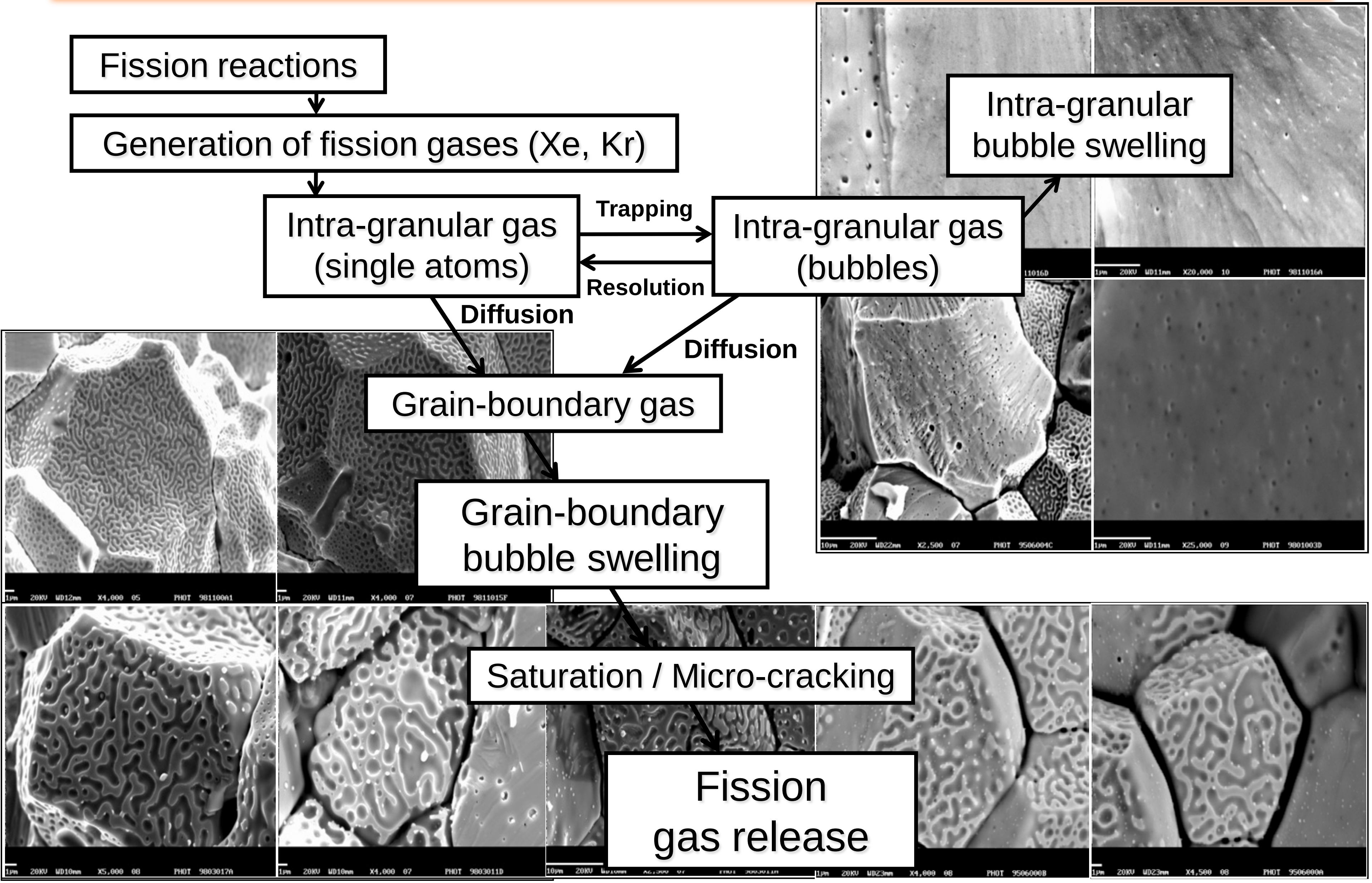
*Presented at SciDAC-4 PI Meeting, 24 July 2018*

# *The long-standing fission gas problem*



Fission reactions

Generation of fission gases (Xe, Kr)

Intra-granular gas (single atoms)

**Trapping**

**Resolution**

Intra-granular gas (bubbles)

Intra-granular bubble swelling

**Diffusion**

**Diffusion**

Grain-boundary gas

Grain-boundary bubble swelling

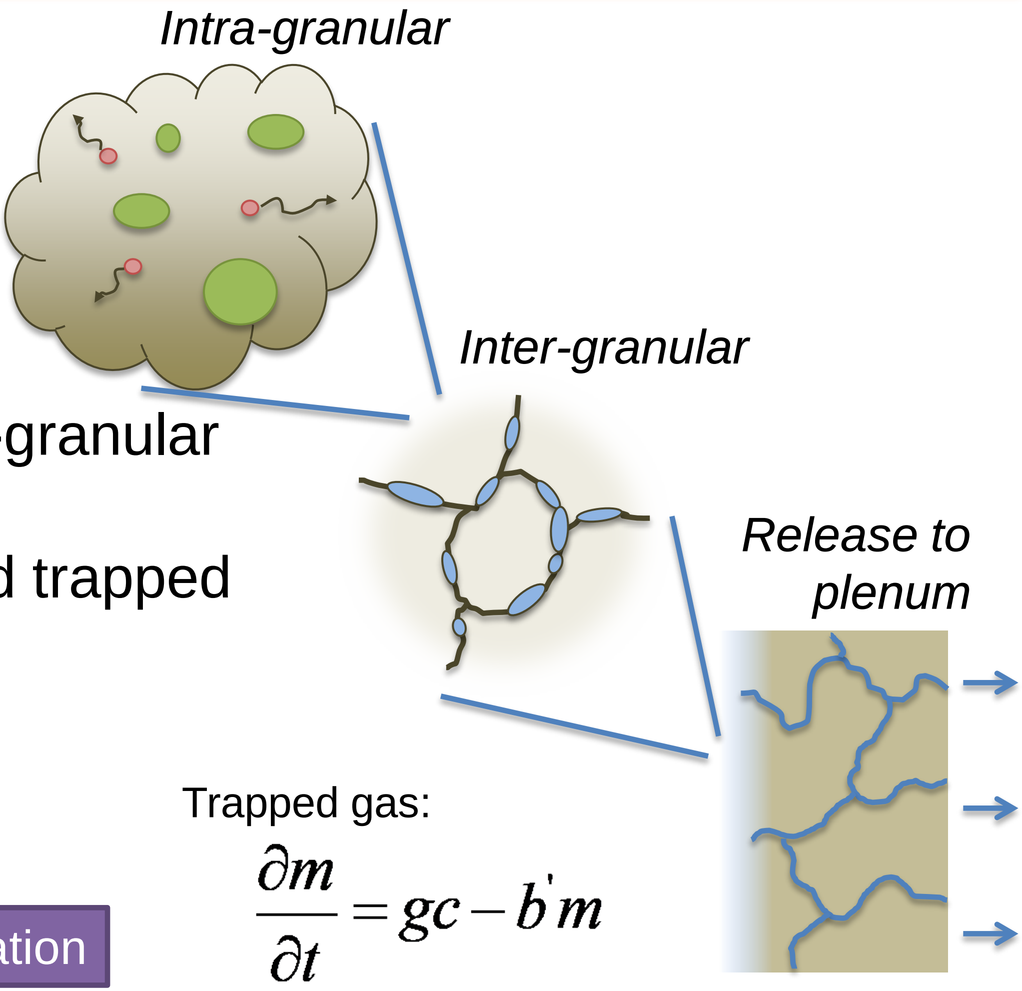
Saturation / Micro-cracking

Fission gas release

* 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:

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

Diffusion

Absorption

Re-solution

Creation

Trapped gas:

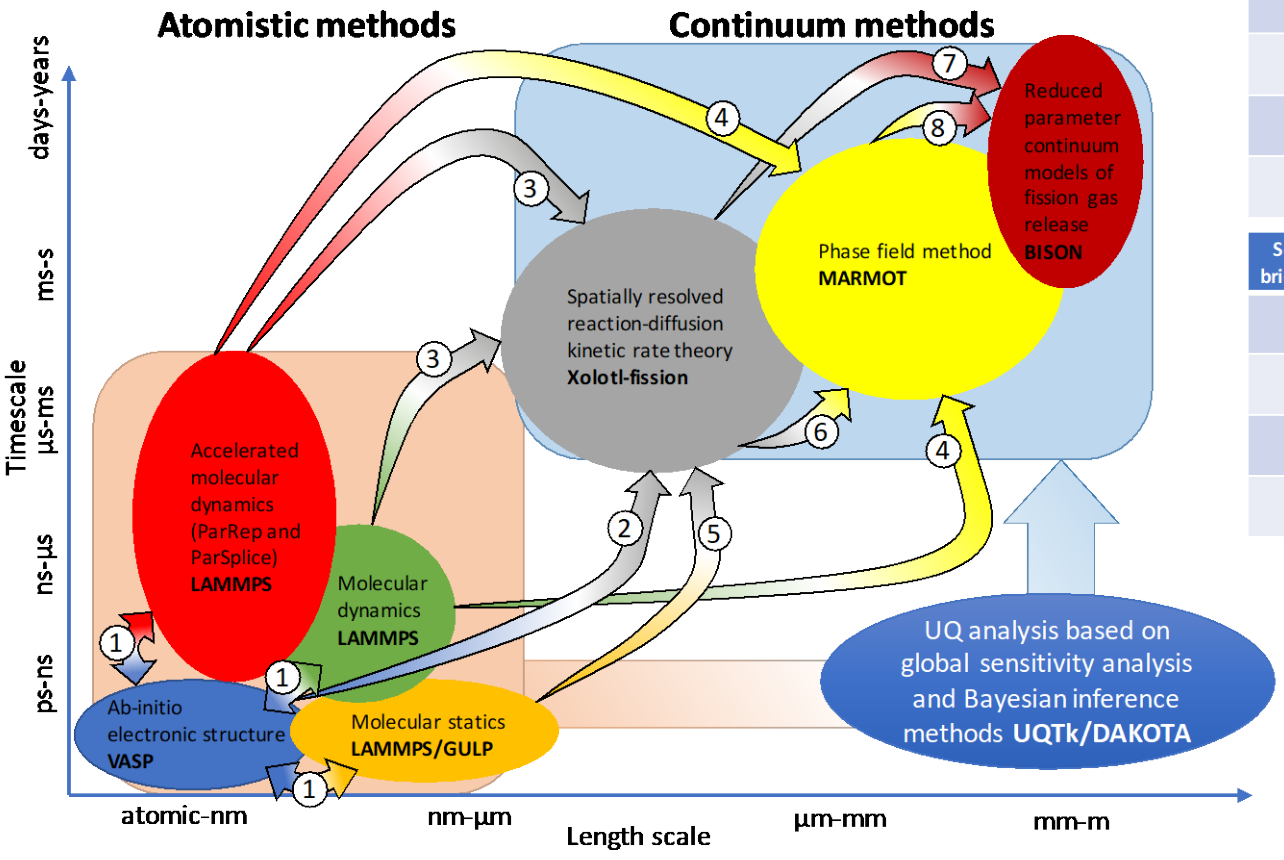
$$\frac{\partial m}{\partial t} = gc - b'm$$

- Effective diffusion rate for accumulation at boundaries:

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

T. Matthews and D. Andersson (LANL)

# *Our vision of multiscale modeling and objectives*



| Scale bridging | Property/information exchanged between scales |
|---|---|
| 1 | Importance of the $Xe_{U6O8}$ and $Xe_{U8Oy}$ clusters, providing a starting point for the AMD simulations. |
| 2 | Migration/formation enthalpies of Xe-vacancy clusters, small bubbles and point defects. |
| 3 | Kinetic properties of Xe-vacancy clusters and point defects. Defect reaction volumes and rates. Bubble resolution. |
| 4 | Kinetic properties of Xe and point defects at grain boundaries/edges and dislocations. |

| Scale bridging | Property/information exchanged between scales |
|---|---|
| 5 | Migration/formation entropies of Xe-vacancy clusters, small bubbles and point defects. |
| 6 | Effective Xe and defect intra-granular diffusion rates. Initial bubble micro-structures. |
| 7 | Effective Xe intra-granular diffusion rates. Intra-granular bubble distributions. |
| 8 | Inter-granular bubble distributions. Thresholds and conditions for release from grain boundaries and edges. |

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*

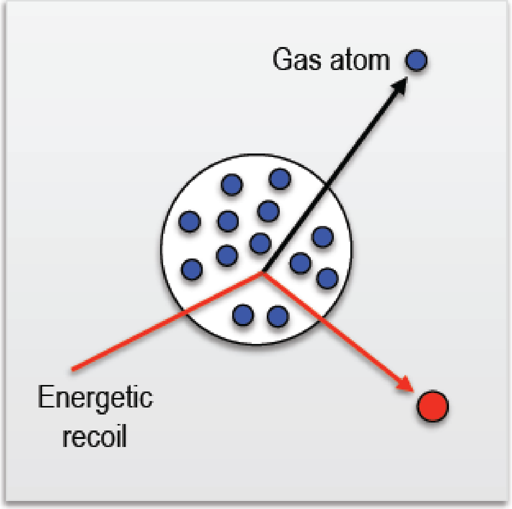
**Computing, Computer Science and Applied Math (David Bernholdt)**

- **Thrust 1 (Blas Uberuaga): 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 (Blas Uberuaga)
- **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)

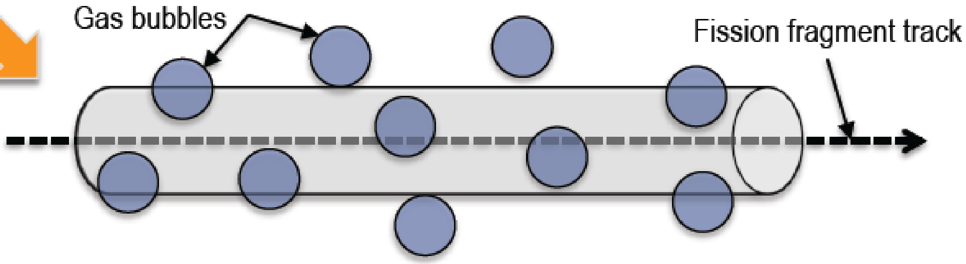
# Pilot project: Large-scale MD simulations of re-solution

Additional Pilot partner: Pacific Northwest NATIONAL LABORATORY Proudly Operated by Battelle Since 1965 W. Setyawan, *et al.*, (2018).

- In homogeneous re-solution, individual gas atoms are ejected from the gas bubble through collisions with energetic fission fragments or recoil uranium atoms that are traversing the bubble
- The kinetic energy acquired by gas atoms through these collisions can range up to the maximum ballistic energy of a fission fragment
- For re-solution to be achieved a gas atom must acquire energy above a critical value $E_{min}$
- In the heterogeneous model bubbles are almost completely destroyed by the passage of a fission fragment in the vicinity of a bubble
- The destruction of the bubble occurs through either the vaporization of the dense gas due to the passing pressure wave or to the trapping of gas atoms in material thrown from one side of the bubble to the other
- This mechanism requires modeling of the entire interaction of the bubble with the fission thermal spike



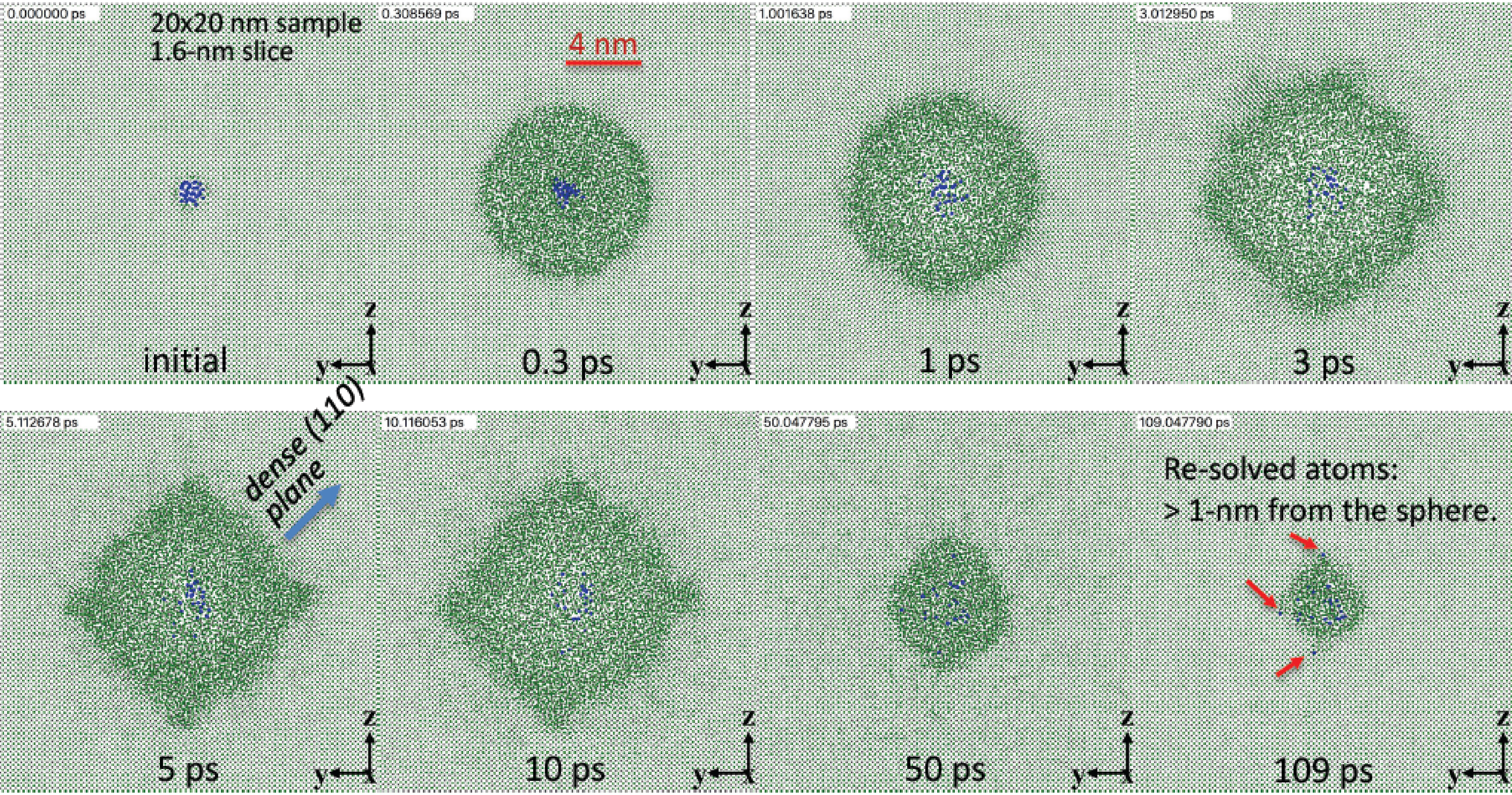
$$b_{hom} = \mu_{ff}\dot{F}\frac{2\pi Z^4 e^2}{E_{ff}^{max} E_{min}} \ln\left(\frac{E_{ff}^{max}}{E_{min}}\right) = \sim 4 \times 10^{-6}\ /s$$



$$b_{het} = \pi\left(R_b + R_{ff}\right)^2\left(2\dot{F}\mu_{ff}\right) = \sim 1.5 \times 10^{-3}\ /s$$

# *MD simulations of heterogeneous resolution*

R0.8N36 bubble subjected to 16 keV/nm thermal spike.

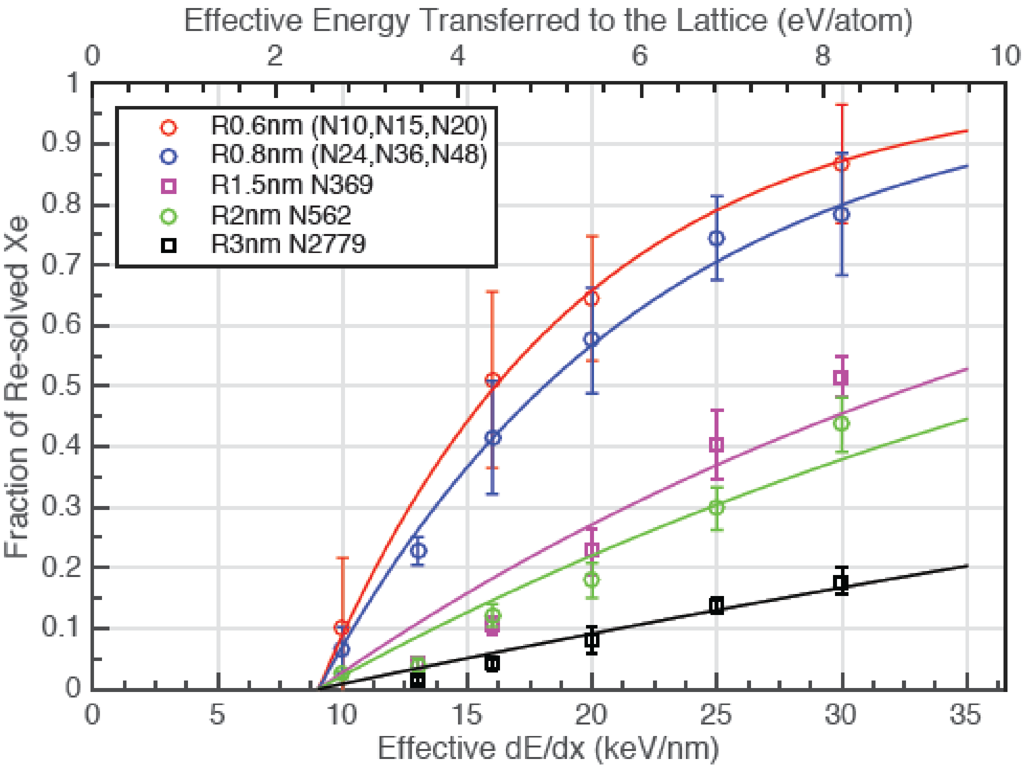


Simulations performed at 600 K with the CRG potential set for $UO_2$+Xe.

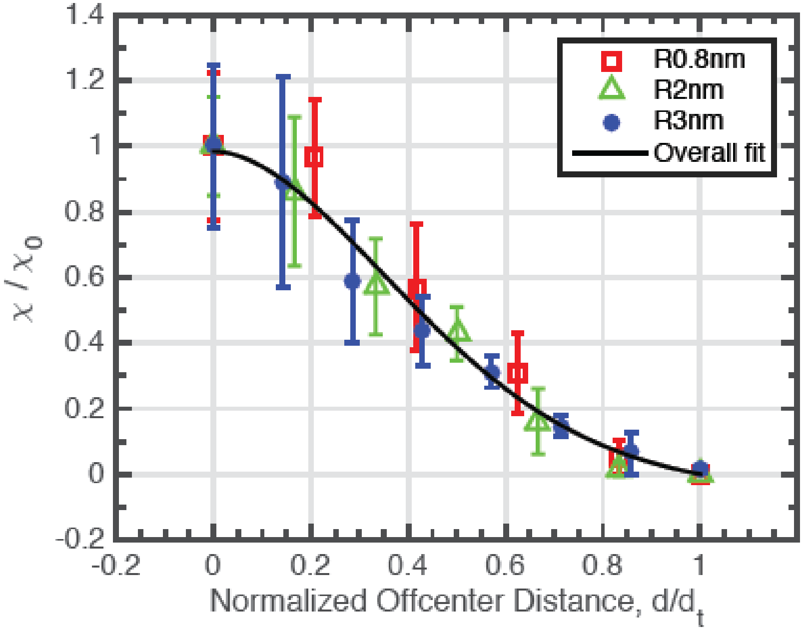
[1] W. D. Cooper, et al., *J. Phys.: Condens. Matt.* **26**, 105401 (2014).
[2] W. D. Cooper, et al., *J. Phys.: Condens. Matt.* **28**, 405401 (2016).
[3] K. T. Tang and J. P. Toennies, *J. Chem. Phys.* **118**, 4976 (2003).
[4] based on e-ph coupling constant $\lambda$ = 4 nm from Toulemonde et al, NIMB 166 (2000) 903.

# Fission gas re-solution mechanisms and new model

On-center Data



Off-center Efficiency



$$\eta \equiv \frac{\int_0^1 y(2\pi x)dx}{\int_0^1 (2\pi x)dx} \approx 0.25$$

$$N_{res} = 0.25N\{1\text{-}\exp[-0.1967*(S_{eff}-9.042)*\exp(-1.1756R)]\}$$

**New model for re-solution derived from the large-scale atomistic simulations:**

$$b_{het} \equiv \pi(R_b + R_f)^2(2\dot{F}\mu_f) * \frac{1}{\mu_f}\int_0^{\mu_f} 0.25\{1 - exp[-0.1967(S_{eff} - 9.042)e^{-1.1756Rb}]\}dx$$

for $S_{eff} > 9.042$ keV/nm, else the integrant = 0.

↑ depend on x

# *Thrust 1: DFT in multiscale modeling needed for Xe diffusion*

Current empirical model:

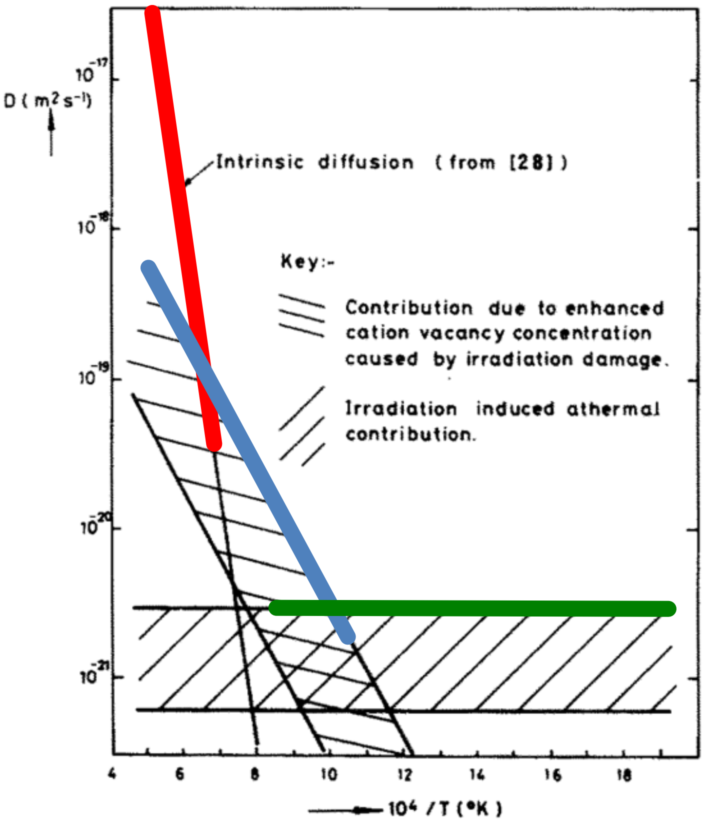
Total: $D_{xe} = D_1 + D_2 + D_3$

Intrinsic: $D_1 = 7.6 \cdot 10^{-10} \times \exp\left(-3.04 / k_B T\right) \left[\mathrm{m}^2/\mathrm{s}\right]$

Irr. Enhanced: $D_2 = 4 \times 1.4 \cdot 10^{-25} \times \sqrt{\dot{F}} \exp\left(-1.2 / k_B T\right) \left[\mathrm{m}^2/\mathrm{s}\right]$

Athermal: $D_3 = 4 \times 2 \cdot 10^{-40} \times \dot{F} \left[\mathrm{m}^2/\mathrm{s}\right]$

- Empirical relationships.
- The mechanisms for $D_1$, $D_2$, and $D_3$ are not understood, which complicates development of predictive models.
- $D_1$ and $D_2$ driven by vacancy population.
  - Diffusion by extended Xe-vacancy clusters.
- $D_3$ is believed to be caused directly by damage.
  - Thermal spikes similar to the bubble re-solution mechanism.

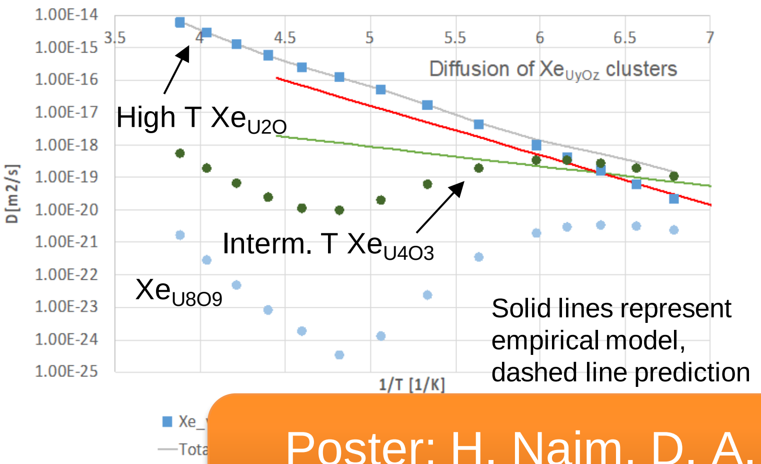


J. A. Turnbull et al., JNM **107,** 168 (1982)

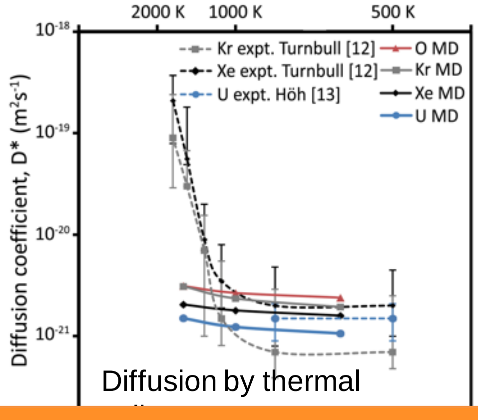
Calculate $D_1$ and $D_2$ fission gas diffusion through simulation using ***point defect dynamics*** and $D_3$ by direct MD simulations.

# Thrust 1 and 3: UQ framework for FG diffusion in $UO_2$

Predictions based on DFT and MD simulations of large Xe-vacancy clusters and thermal spikes

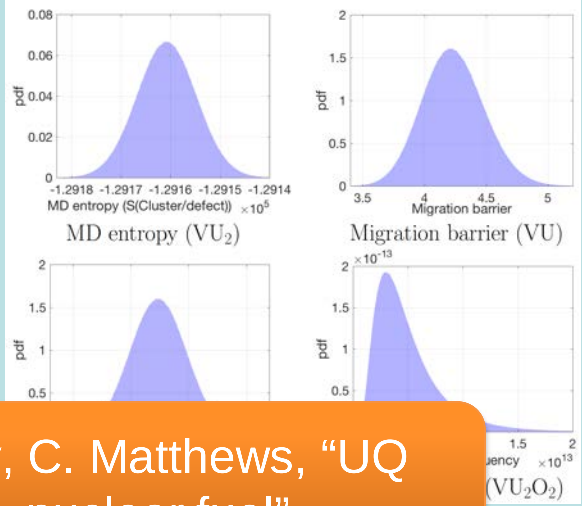


Solid lines represent empirical model, dashed line prediction



Diffusion by thermal

## Input parameter PDFs for UQ analysis



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

Poster: H. Najm, D. A. Andersson, T. A. Casey, C. Matthews, "UQ framework for fission gas behavior in $UO_2$ nuclear fuel"

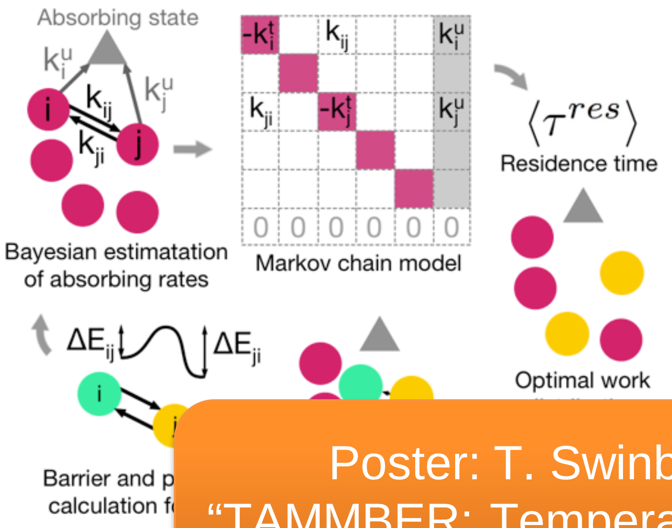
- Desire estimates of confidence in predictions and identification of primary sources of uncertainty with respect to quantities of interest (QoIs).
- UQ approach: Deploy statistical methodologies to efficiently represent mappings from uncertain model inputs to uncertain outputs (propagation), identifying critical input parameters that affect QoI uncertainty (sensitivity), constructing robust representations of input parameter uncertainty as informed form available data (statistical inference).

# *Thrust 1: TAMMBER (AMD simulations of defect clusters)*

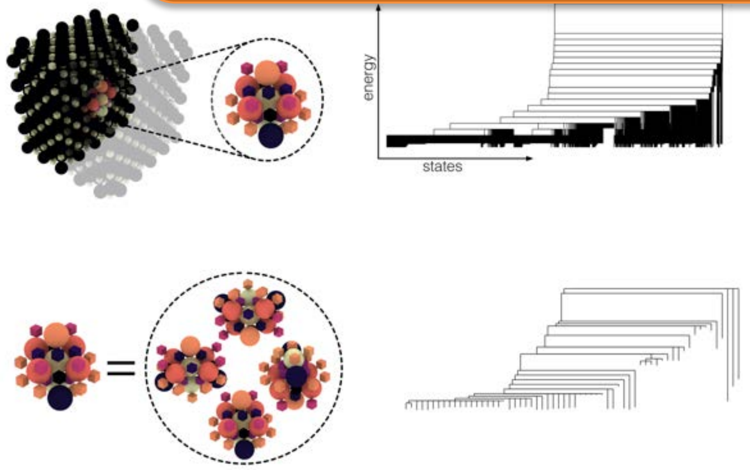
- **TAMMBER:** Temperature Accelerated Markov Models with Bayesian Estimation of Rates (*T. Swinburne, et al., Phys. Rev. Mater, 2018*).
- **Autonomous management is essential to fully harness modern supercomputers:**
  - Due to the rate of data production, user supervision is a critical bottleneck when building mesoscale material models from

parameterization of AMD to reduce model uncertainty and maximize prediction timescale.

- Massively parallel distribution of TAD simulations autonomously builds a transition network to predict seconds of material evolution with uncertainty quantification.
- Determination of the full rate matrix for a defect, appropriate for upscaling to mesoscale codes.



Poster: T. Swinburne, et al. (presented by B.P. Uberuaga), “TAMMBER: Temperature Accelerated construction of Markov Models with Bayesian Estimation of Rates” and “Applications of TAMMBER”

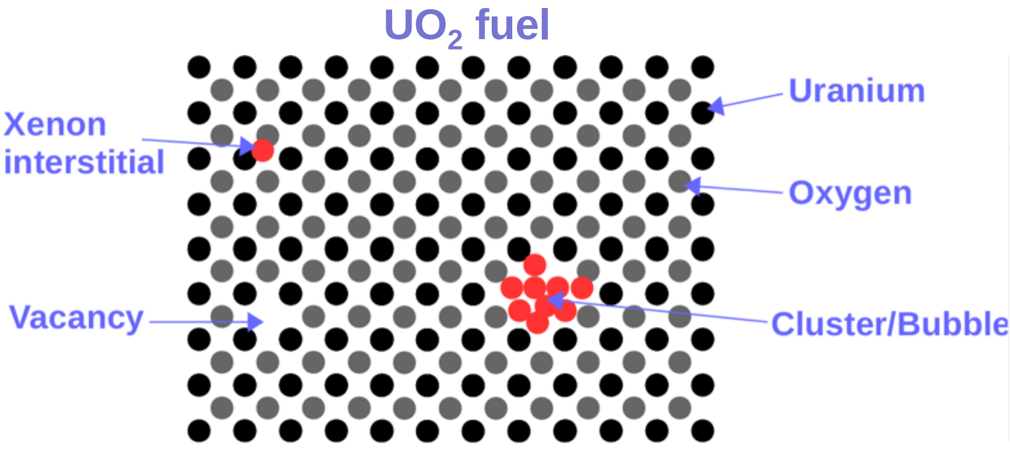


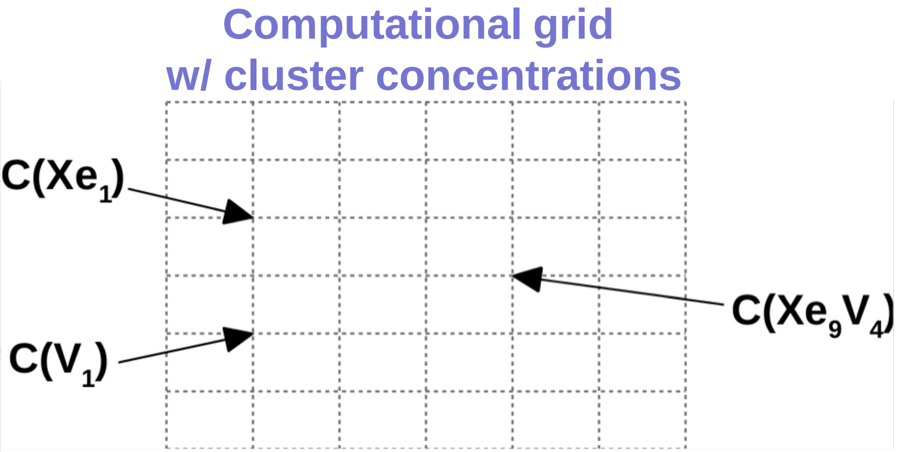
Application to different defects in $UO_2$

# Thrust 2: 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/

# *Xolotl: Design, implementation and challenges*

**Core dependencies:** C++, MPI, HDF5, PETSc

**Key features**

- Built around PETSc solver (FASTMath/ANL collaboration).
- Built-in always-on performance monitoring (SUPER/RAPIDS/ORNL collaboration).
- In situ visualization of data and performance (SDM/ORNL collaboration).
    - Initially using EAVL now migrated to VTk-m.
- Performance has not been a major issue (starting to change):
    - Ongoing incremental performance improvements as bottlenecks identified (SUPER/RAPIDS/ORNL collaboration).
    - Beginning to implement acceleration for many-core and GPU.
    - Performance and resource usage characteristics differ between fission and fusion use cases due to very different reaction networks.
        - Memory requirements are a major issue for fusion.

**SciDAC-4 challenges**

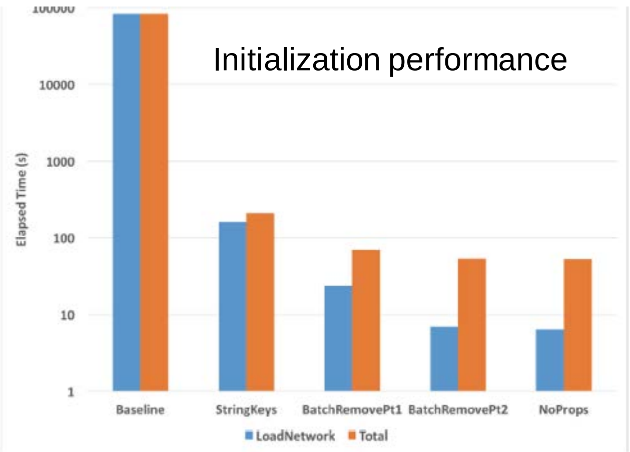
- Scaling up grain size:
    - Both memory and performance issues expected.
- Coupling with MARMOT:
    - Primarily uncertainties due to new territory.

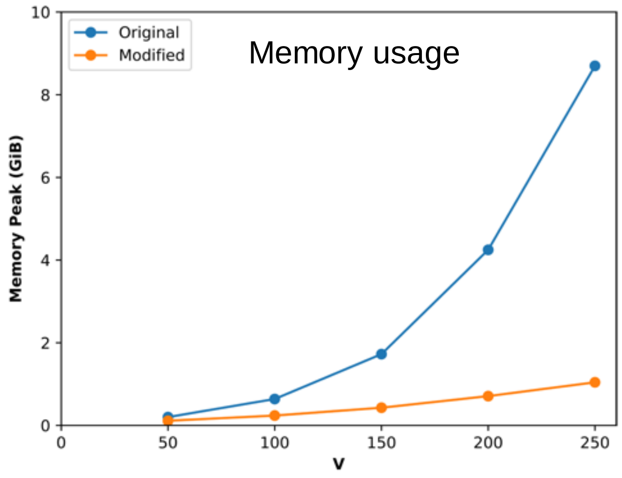
# *Xolotl: Improving efficiency and initial results*

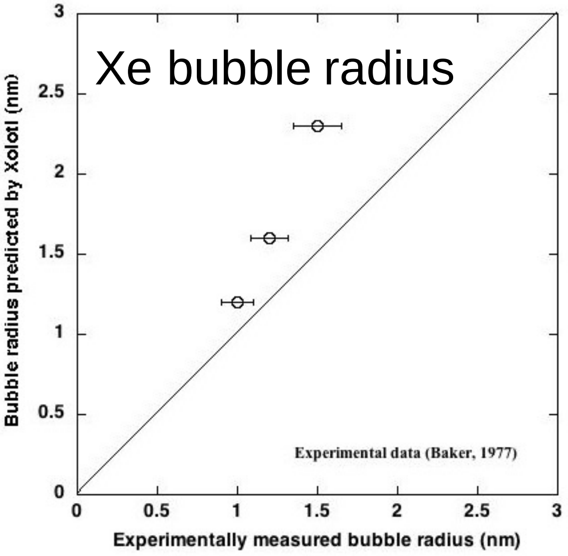
- The initialization of the Xolotl reaction network did not scale well with respect to ($Xe_n$,$Va_m$):
  - Test problem size was intractable due to poor initialization performance.
  - Diagnosed performance using Xolotl’s built-in tools and implemented remedies.
- Grouping schemes are used to reduce the number of degrees of freedom while modeling the same physics:
  - Still resulted in high memory usage, profile Xolotl to reduce the usage.
  - I
    S
- Impl nabled first simulation of intragranular gas bubble evolution (without re-solution at this point):
  - Slight overestimation compared to experiments due to lack of re-solution.

Poster: Sophie Blondel, Phil Roth, David Bernoldt, David Andersson, Brian Wirth, “Xolotl: a cluster dynamics code to predict gas bubble evolution in solids”

All tests on OLCF EOS system







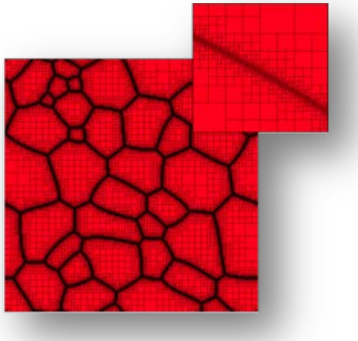
# *Thrust 2: MARMOT meso-scale fuel performance tool*

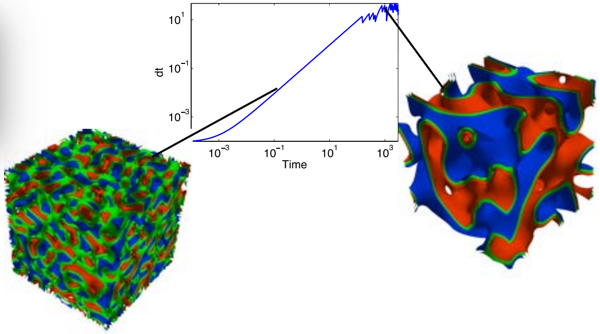
- **Predicts the coevolution of microstructure and properties in nuclear materials.**

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

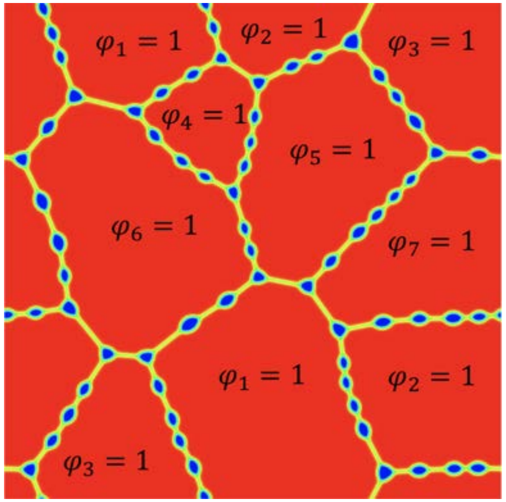
**MARMOT:**

- 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



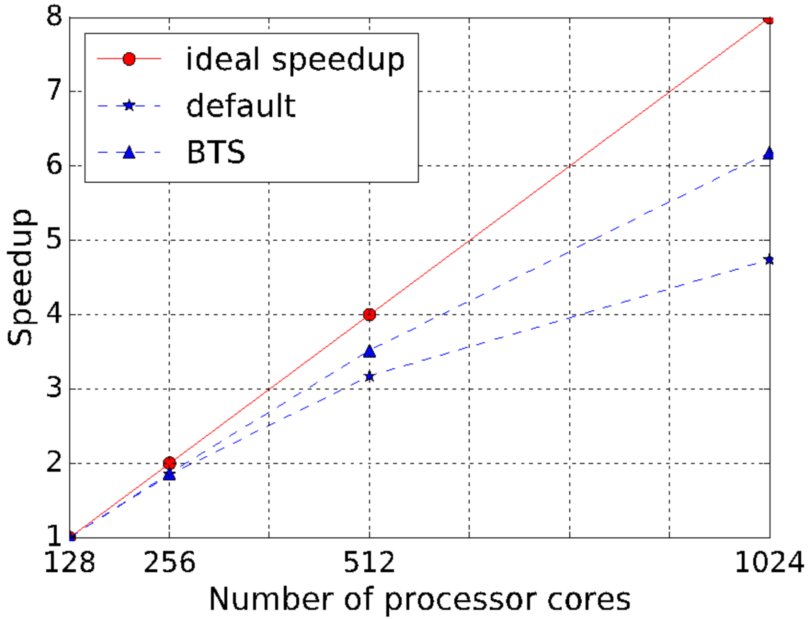


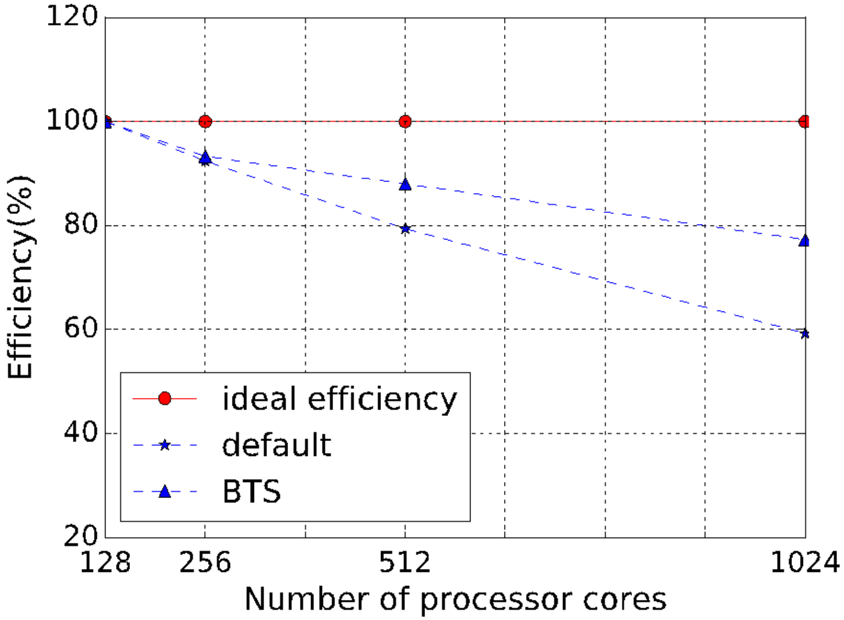
- **MARMOT fission gas model predicts:**
  - **Transport of gas atoms and U vacancies (2 DOF).**
  - **Void growth and coalescence (1 DOF).**
  - **Grain boundary migration (1 – 20 DOF).**
- **The model uses 4 – 22 DOF per node.**



# *MARMOT numerical improvements needed*

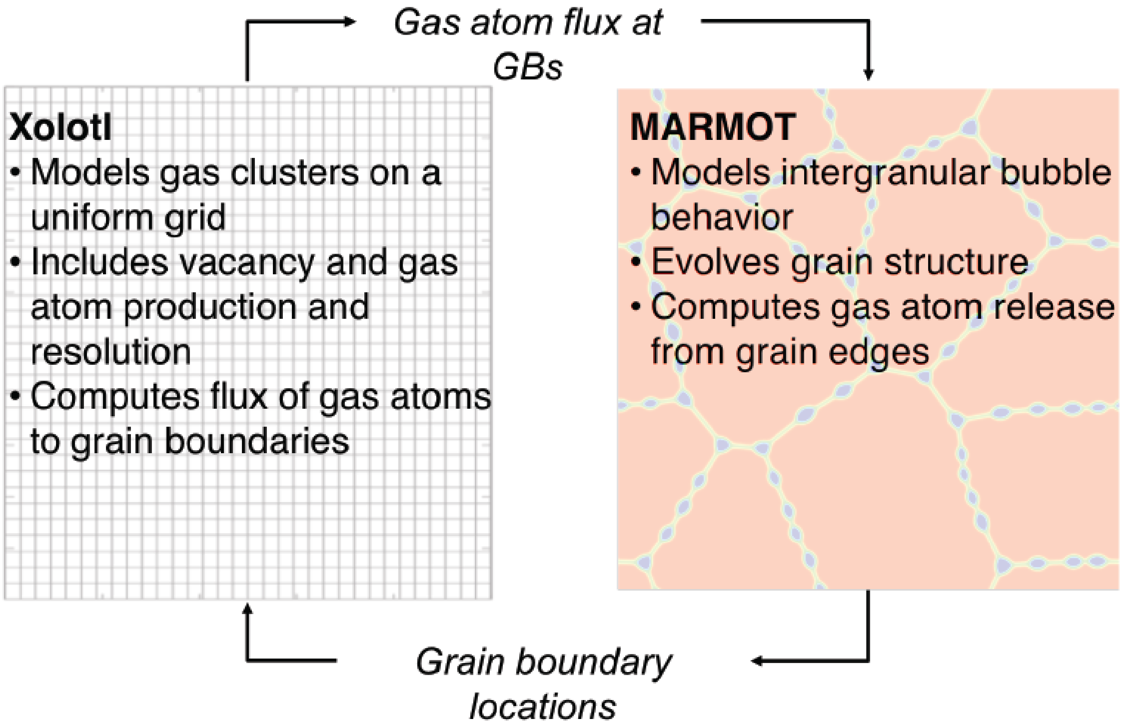
- **MARMOT fission gas simulations will require O(30,000) processors**
  - **It is typically run on 10 – 1000 processors**





  - **Preliminary scaling study solved 3D problem with 22,180,149 DOF:**
    - Compared the default vector communication settings in the PETSc library with a new method denoted “BTS”.
  - **Improvements needed in MARMOT for large runs:**
    - Communication (BTS is better but still room for improvement).
    - Partitioning.
    - Memory usage.

# Xolotl and MARMOT coupling approach



*Gas atom flux at GBs*

**Xolotl**
- Models gas clusters on a uniform grid
- Includes vacancy and gas atom production and resolution
- Computes flux of gas atoms to grain boundaries

**MARMOT**
- Models intergranular bubble behavior
- Evolves grain structure
- Computes gas atom release from grain edges

*Grain boundary locations*

- **Coupling will be managed by the multi-app system in MOOSE.**
- **The codes will each converge separately and then pass information.**
- **Making the coupling scale well is of primary importance.**

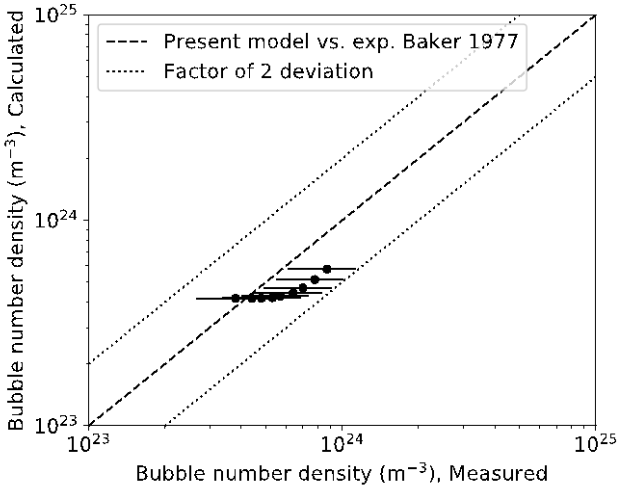
# *Thrust 2 and 3: Reduced order model in BISON and experimental validation*

- BISON is a fuel performance code based on the MOOSE framework developed at INL.
- We have implemented a new engineering scale model for fission gas bubble density, concentration of gas in the bubbles and concentration of gas dispersed in the matrix.
- Destination for results from coupled Xolotl-MARMOT simulations.

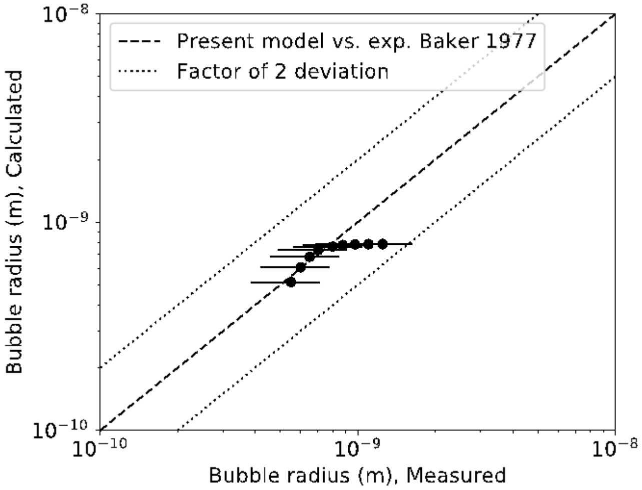
Governing equations:

$$\frac{\partial N}{\partial t} = +\nu - \alpha_{\bar{n}} N \qquad \frac{\partial \psi}{\partial t} = +2\nu + \beta_{\bar{n}} N - \alpha_{\bar{n}} \psi \qquad \frac{\partial c_1}{\partial t} = +yF + D\nabla^2 c_1 - 2\nu - \beta_{\bar{n}} N + \alpha_{\bar{n}} \psi$$

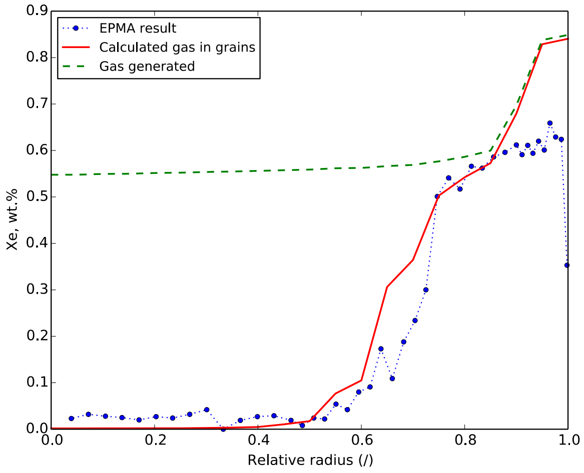
Predicted vs measured bubble density



Predicted vs measured bubble radius

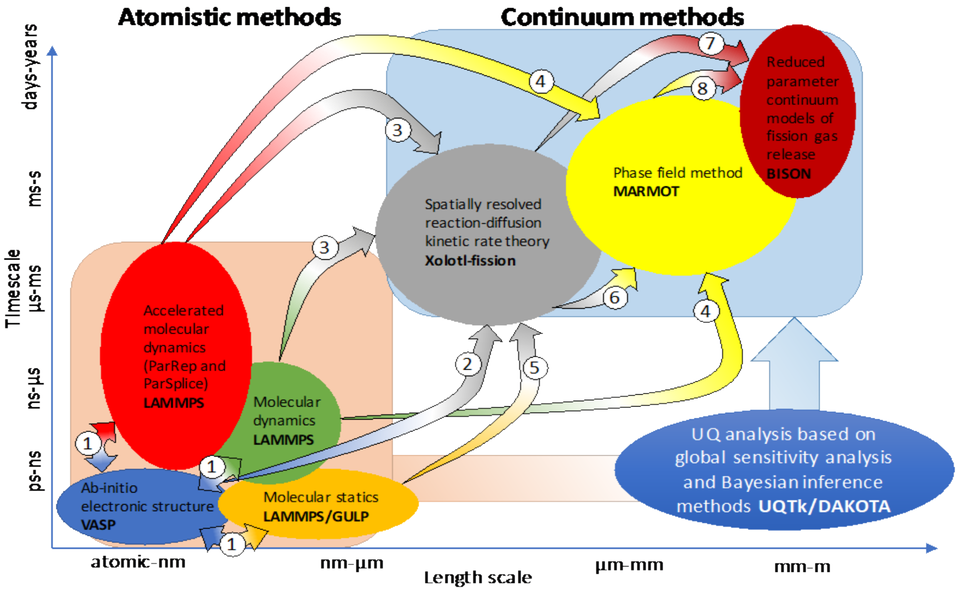


Measured vs simulated Xe concentration as function of radial position



# *Thrust 3: UQ plans, targets and work in progress*

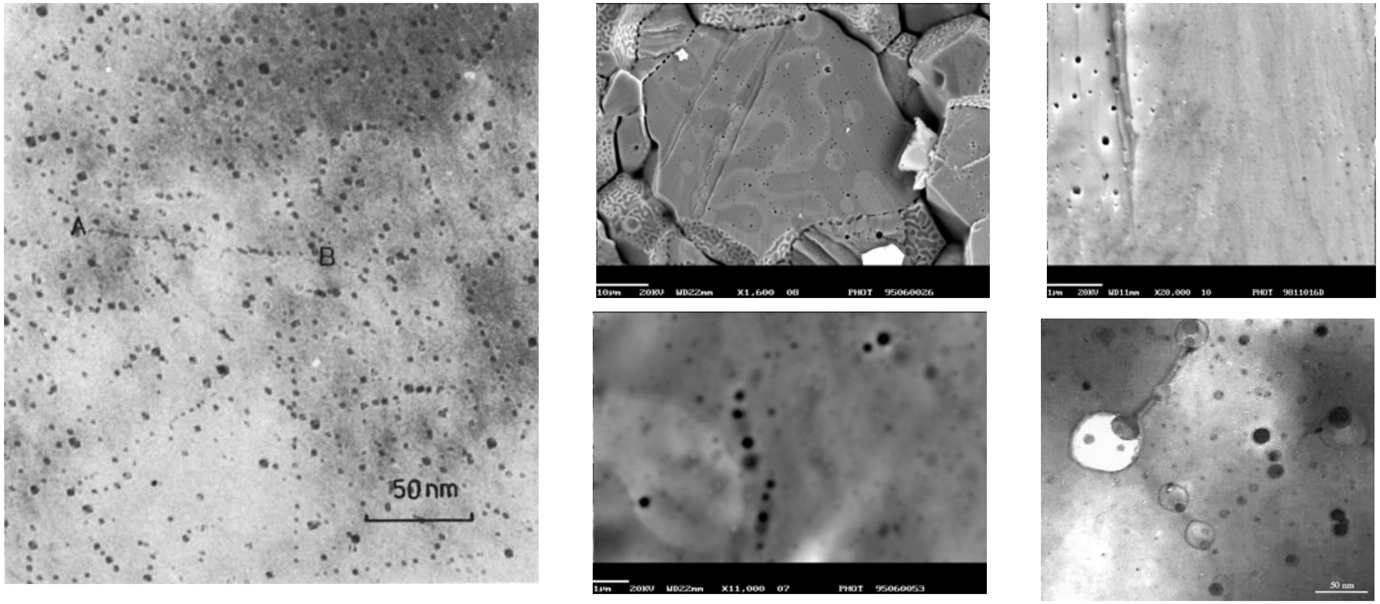
- We are pursuing UQ in the coupled multiscale suite of models:
  - Tightly coupled continuum modeling with MARMOT & Xolotl.
  - Uncertain atomistic simulations, as well as exptl measurements, inform MARMOT-Xolotl.
  - Uncertain MARMOT-Xolotl predictions inform BISON.



- Thrust activities span:
  - Global sensitivity analysis (GSA).
  - Forward uncertainty propagation.
  - Bayesian inference and parameter estimation.
  - Model validation with experimental data.
- UQ work in progress:
  - GSA for DFT modeling, to be followed by forward UQ, Bayesian inference calibration, estimate uncertain predictions and Bayesian model selection.
  - GSA for MARMOT+Xolotl, to be followed by forward UQ, estimate uncertain predictions, provide inputs to BISON.

# Thrust 3: Experimental validation

- Extensive experimental database being considered that includes:
  - Irradiation tests at normal operating temperatures (Baker, J. Nucl. Mater. 66, 1977) and power ramps and cycles (White, R&T/NG/EXT/REP/0206/02, 2006).
  - Post-irradiation annealing tests (Kashibe, JNM 206, 1992).
  - Integral fuel rod tests (Mogensen, JNM 131, 1985).



*TEM micrograph from Baker (left) and SEM micrographs from White (right) showing intra-granular fission gas bubbles in irradiated* $UO_2$

- Experimental validation is ongoing for each tool at their specific scale, the next target is validation in the multi-scale domain.
- Coordinated effort with UQ in order to assess predictivity in the light of uncertainties.

# *Summary and conclusions*

- The 3 stages of fission gas release addressed by a multi-scale approach:
  - Atomic scale simulations to understand the rates and stabilities of point defects interacting with Xe atoms.
  - Meso-scale modeling of gas evolution within grains using the Xolotl-fission cluster dynamics code and intergranular evolution using the MARMOT phase field code, to be coupled in the present project.
  - Engineering scale simulations of gas release using the BISON code.
  - Full integration of uncertainty quantification and experimental validation.
- Computer scientists enable us to deploy tools on leadership-class computers.
- Initial results highlight:
  - New bubble resolution model.
  - Diffusion mechanism under irradiation remains uncertain, but likely involves large clusters. Conclusions are analyzed by UQ of the model parameters.
  - TAMMBER AMD tool for long-time scale simulations with Bayesian estimation of rates.
  - Performance assessments/improvements of Xolotl-fission and MARMOT, with results demonstrating capabilities and a coupling approach.
  - New engineering scale model in BISON assessed against experiments.