

Yearly Progress Report (June 2018):
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Title: Simulation of Fission Gas in Uranium Oxide Nuclear Fuel

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The behavior of xenon (Xe) in nuclear fuel is of critical importance to nuclear fuel performance, because the diffusion and precipitation of Xe in fission gas bubbles influences both the amount of fuel swelling and the quantity of fission gas released to the fuel rod plenum [1]. Despite decades of investigation, significant uncertainties exist regarding the underlying mechanisms controlling Xe diffusion, precipitation and release that impact predictions of fission gas swelling and release during both normal operation and transient conditions in accidents. Such uncertainties limit the accuracy of integrated fuel performance models [2], which are important to many DOE Office of Nuclear Engineering programs including the Nuclear Engineering Advanced Modeling and Simulation (NEAMS) and the Consortium for the Advanced Simulation of Light water reactors (CASL). Most fission gas behavior models used for fuel performance calculations trace back to the 1957 formulation by Booth [3], the mid-to-late 1960's formulation by Speight and co-workers [4,5]; or the slightly more recent work by Turnbull [6], White and Tucker [7], and Forsberg and Massih [8,9]. These models typically rely on a small number of spatially independent partial differential equations, or even a single partial differential equation, along with the concept of an effective diffusivity to account for the effect of intra-granular fission gas bubbles on the bulk diffusion rate [4]. These models are clearly over-simplifications of the actual behavior of noble gas within the fuel. Indeed, the dynamics of fission gas bubbles, which form both within the grains (intra-granular) and on grain boundaries (inter-granular), affects the rate of fission gas diffusion and release, and also determines the fission gas swelling [1, 7,10]. Despite being a key determinant of fission gas effects, which control fuel performance, accurate physically based models of intra-granular bubble evolution are still lacking in current models. Rather, the characteristics of the bubble population (number density, mean size) are given as constant parameters or calculated through simplistic, empirically based functions of temperature [2,7,8]. As the complex bubble evolution process strongly depends on the specific fuel irradiation conditions and reactor operating history [11-13], this gap in current modeling capability significantly limits the accuracy and transferability of available treatments. Therefore, developing effective modeling capabilities for fission gas bubble evolution is a key issue in achieving higher standards of accuracy for predictions of fission gas swelling and release in fuel performance analysis.

The objective of this project is to advance the mechanistic understanding of fission gas behavior in UO₂ nuclear fuel by developing a mesoscale fission gas simulator that takes advantage of leadership class computers. This will enable an accurate and physically based fission gas release model for application in integrated fuel performance codes. The model will be informed by results from large-scale atomistic and mesoscale simulations of the three stages of fission gas release, including diffusion and intra-granular bubble formation, bubble growth and coalescence on grain faces, and the transport of gas through interconnected grain edge tunnels to the fuel rod plenum. In order to address this complex problem,

existing atomistic and mesoscale simulation tools will be further developed to take advantage of DOE high performance computing capabilities. The simulation tools will undergo rigorous uncertainty quantification and validation. In addition to gas release and retention, the integrated tool will predict their impact on performance parameters such as fuel swelling and thermal conductivity.

Our project involves developing an advanced mechanistic model of fission gas release in oxide nuclear reactor fuel that consider all three stages of gas release and explicitly includes the impact of both intra- and inter-granular gas bubbles. The development of this model will use the insights from molecular dynamics (MD) simulations with accelerated MD techniques (AMD) and selected density functional theory (DFT) calculations to provide quantitative assessments of fission gas diffusion. The behavior of the intra-granular bubbles will be investigated using cluster dynamics. We will rely on a hybrid, and spatially-dependent, continuum model, containing discrete cluster dynamics for clusters of size up to a few hundred gas atoms and a grouping scheme bundling a range of cluster sizes into distributions for capturing larger clusters. The inter-granular bubble behavior will be investigated using the phase field method (PFM) by enhancing the numerical performance of the MARMOT phase field code used to solve the problem. Both the cluster dynamics and phase field simulations will be deployed on DOE HPC systems. The cluster dynamics code focusing on intra-granular fission gas behavior will be coupled to phase field simulations by providing the flux of gas atoms to the grain boundaries. Our project also involves the use of uncertainty quantification across the simulation length and timescales, and will validate predictions against existing and emerging data on fission gas bubble distribution in nuclear fuel. As well, the uncertainty of the final fission gas release model will be quantified and compared against the uncertainty of the existing data to ensure that our development efforts have significantly decreased the uncertainty and increased the accuracy of the engineering-scale fission gas release model.

The outcome of this project will be improved insight into the atomistic processes of Xe diffusion and clustering, and the validation of a leadership class fission gas simulator, Xolotl-fission, for modeling the spatially dependent, and multi-modal intragranular bubble population evolution within nuclear fuel. Further, the Xolotl-fission code will be coupled to the mesoscale MARMOT phase field model to provide a self-consistent, high fidelity modeling approach for both intragranular and intergranular fission gas bubbles, leading to an improved physical model of fission gas release. These outcomes will set the stage for future multiscale model research activities, coupled to a comprehensive uncertainty quantification analysis, to ultimately treat fission gas swelling and release as inherently coupled to bubble evolution. Such advances are crucially needed to deliver improved, physics-based models needed by advanced, engineering-scale fuel performance models under development within the DOE NE programs, namely the CASL and NEAMS.

In order to accomplish these goals, the research is organized in three thrusts with defined interfaces between them:

Thrust 1: DFT and long-time scale atomistic simulations to understand fission gas and defect behavior. Fission gas diffusion is enabled by uranium and oxygen vacancies. We are using molecular dynamics (MD) and in particular accelerated molecular dynamics (AMD) techniques to directly simulate the kinetic behavior of large Xe-vacancy clusters and their interactions with other defects. The MD and AMD simulations are complemented by selected DFT calculations. These properties are critical for an accurate mesoscale description of fission gas evolution. The thermodynamic and kinetic properties of fission gas atoms at grain boundaries and dislocations will also be probed by MD and AMD methods in order to provide data for the phase field model of inter-granular bubbles.

Thrust 2: Spatially discretized cluster dynamics and MARMOT phase field simulations to understand fission gas bubble behavior. This task involves two continuum approaches tailored to describe 1) nucleation, growth and re-resolution of intra-granular bubbles and 2) growth and coalescence of inter-granular bubbles on grain faces and edges. The first approach is a hybrid and spatially dependent continuum model, called Xolotl-fission, consisting of spatially-discretized, 3D reaction-diffusion cluster dynamics coupled to a methodology to treat the larger size bubble population evolution to model strongly bi- (or, in some cases multi-) modal bubble size distributions. The two methods will be coupled as Xolotl-fission will provide fluxes of gas atoms to grain boundaries to a phase field model (the second approach), as implemented in the mesoscale MARMOT code to capture the growth and interconnection of grain face bubbles and how they interact with grain edges to form tunnels that allow gas atoms to vent to free

surfaces in massive 3D polycrystal simulations. MARMOT developers working with ASCR funded scientists are improving the computational efficiency of MARMOT to facilitate these 3D microstructures requiring leadership class computational resources. Both Xolotl-fission and MARMOT will be used to improve the understanding of fission gas behavior and release, and support development of a reduced order fission gas release model for application to the BISON fuel performance code.

Thrust 3: Uncertainty quantification and experimental validation. Global sensitivity analysis is used to identify important fission gas diffusion mechanisms. These will be applied to both the Xolotl-fission and MARMOT as well as to the coupled version of the codes, to assess the impact of specific gas diffusion mechanisms on intra-granular bubble evolution, and subsequently inter-granular bubble evolution and fission gas release. We will also apply Bayesian inference methods for statistical calibration of fission gas diffusion models with embedded model error representations, arriving at probabilistic representations of relevant uncertain model parameters. These will subsequently be used in Xolotl-fission and MARMOT computations, providing predictions of fission gas bubble populations and diffusion coefficients with quantified uncertainty. Available data will be used to assess confidence in the underlying models, via Bayesian model evidence estimation, thereby providing means for model validation, hypothesis testing and model selection. Experimental validation will particularly emphasize validation against experimental work on the continuum (microstructure) and engineering scales.

Computationally, the project leverages a great deal from the SciDAC-PSI project [14], with a common computer science/applied math team working on both projects. This enables us to focus the computational work on the areas of difference between fission in nuclear fuel and plasma-surface interactions. The project has established a web presence at <https://collab.ceels.anl.gov/display/FissionGasSciDAC2>, leveraging R&D infrastructure available in the ANL Mathematics and Computer Sciences Division. This wiki site has both public-facing and project-private spaces. Currently, the public space provides basic information about project goals, participants, sponsors, and key collaborators, along with a catalog of project presentations and publications. The private space includes records of project and working group meetings and other information. Additionally, we use a Subversion repository, also hosted at ANL, as a repository for internal and under-development documents, and the project mailing list is hosted at ORNL.

This document describes the initial progress of the project since the initial kick-off meeting held in December 2017 at the University of Florida. Our most significant outcomes have involved improvements in Xolotl-fission performance, the implementation of a new bubble re-resolution model, based on a systematic set of MD simulations performed during the pilot NE SciDAC project, developing an approach for coupling Xolotl-fission to MARMOT, initiating an approach to uncertainty quantification, and initial work performing accelerated MD simulations.

INITIAL PROGRESS ON PROJECT RESEARCH ACTIVITIES

Computer Science/Applied Math

Work in the computer science and applied mathematics (CS/AM) component of the project has proceeded with activities supporting all of the thrusts. The activities in Thrust 2 have focused on improving the performance of the Xolotl code, and developing an initial approach for coupling Xolotl and MARMOT. Thrust 3 has focused on uncertainty quantification and experimental validation.

Performance and scalability improvements in Xolotl: In order to reach experimental time scales, Xolotl needs to be able to model larger and larger networks of fission gas clusters/bubbles within nuclear fuel grains. However, the current version of Xolotl suffers from a memory usage problem, namely that simulation runs would fail due to out-of-memory errors at smaller network sizes than expected on many high-performance computing (HPC) systems.

As part of our ongoing effort to improve Xolotl performance, we have also been investigating the use of threads on systems that do not contain graphics processing units (GPUs). We recently conducted a study comparing runtime configurations that vary the ratio of threads to processes used in a Xolotl run. Figure

2 shows the time required to complete the most expensive parts of program initialization and time stepping on the OLCF Eos system. We determined that a high threads-to-processes ratio benefits the initialization, but time-stepping does best with a single thread per process in the current implementation. We are currently working on an implementation that changes the number of threads used as the simulation runs.

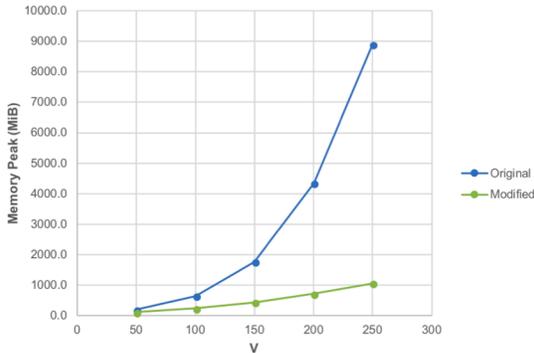


Figure 1. Peak memory usage for original Xolotl implementation and our modified version when running 1D problem on 16 processes in 1 node of the OLCF Eos Cray XC30.

application programming interface (API), we could specify the same information using a sparse matrix representation. As shown in Figure 1, our modification reduced the peak memory requirement by 88% for the largest problem that the original version could run on the Oak Ridge Leadership Computing Facility (OLCF) Eos Cray XC30, namely $\{\text{Gas atom (Xe), } V\} = \{1000, 250\}$. The modification also resulted in a performance benefit, running approximately $1.2\times$ faster than the unmodified code for a smaller problem representing a phase space with a maximum vacancy size of 200. Our addition to the PETSc API has been integrated into the PETSc development repository by Barry Smith (ANL) for inclusion in future releases. Similarly, at each time step Xolotl was using a non-sparse matrix to compute and transfer the Jacobian to the PETSc solver, which is then using a sparse matrix in the solver. This Xolotl matrix has now been reduced to a sparse matrix, which lead to a reduction of memory used from 1.4 GB per core to 230 MB during the time steps for the maximum vacancy size of 200.

As part of our ongoing effort to improve Xolotl performance, we have also been investigating the use of threads on systems that do not incorporate any graphics processing units (GPUs), since our target system does not contain GPUs. We recently conducted a study comparing runtime configurations that vary the ratio of threads to processes used in a Xolotl run. Figure 2 shows the time required to complete the most expensive parts of program initialization and time stepping on the OLCF Eos system. We determined that a high threads-to-processes ratio benefits the initialization, but time-stepping does best with a single thread per process in the current implementation. We are currently working on an implementation that changes the number of threads used as the simulation runs.

Phil Roth (ORNL), working with Sophie Blondel (UTK), explored two different approaches to address this problem: the use of a grouping scheme to gather clusters together and reduce the number of degrees of freedom but still model the same physics, and profiling Xolotl to purely and simply reduce the memory usage. We identified that the high memory consumption of Xolotl resulted from the need to create two very large two-dimensional matrices, which will be required to ultimately model the coupled vacancy – xenon cluster phase space to accurately model fission gas bubble nucleation, used to specify the fill pattern of the sparse matrix to the PETSc solver. We determined that the number of non-zeros in these matrices was small, and that with a minor addition to the PETSc

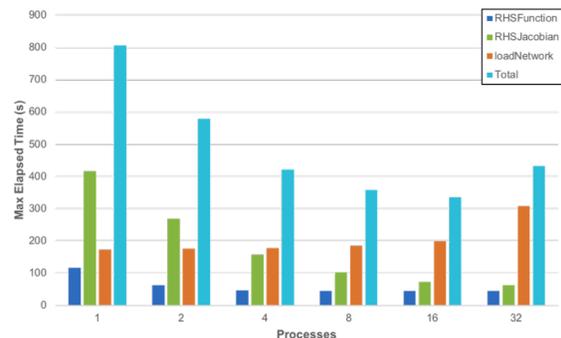


Figure 2. Time required to run the three most expensive activities of a Xolotl simulation run when trading off processes for threads. Timings shown for 1D problem on 32 total threads in 1 OLCF Eos compute node.

MARMOT scaling study: MARMOT uses the phase field method to model the intergranular bubble behavior and grain boundary migration. The MARMOT model is composed of a number of partial differential equations that must be solved simultaneously to describe the microstructure evolution. MARMOT is based on the Multiphysics Object-Oriented Simulation Environment (MOOSE). While MOOSE has powerful high-performance computing capabilities, improvements are needed to effectively use 10,000 or more processors. These improvements will be carried out throughout the project, however the first step is to evaluate the current capabilities in MOOSE. As MARMOT directly builds on MOOSE, it will have the same parallel performance.

In order to evaluate the current parallel scalability of the phase field models in MOOSE, a scaling study is being carried out using a phase field grain growth model that is very similar to the intergranular fission gas model that is still under development. The mesh used in the study has 2,464,461 nodes and 22,180,149 degrees of freedom. The nonlinear system of equations is solved by a Jacobian-free Newton Krylov method together with the additive Schwartz method and the pre-conditioned generalized minimal residual method. We used the default vector communication settings in the PETSc library, as well as a new method denoted “BTS”, where the communication pattern is reused and the messages are compressed before sending. The speedup and efficiency are shown in Figure 3. Within each plot of Fig. 3, the ideal speedup is shown for reference. From these results, it is clear that BTS method uses the processors much more efficiently than the default settings, though these studies are ongoing using 2048 and 4096 processors.

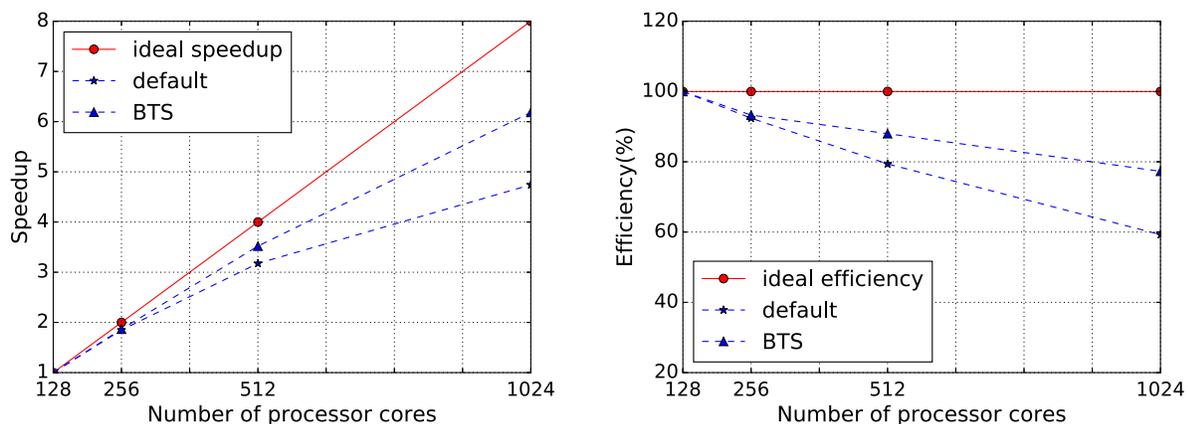


Figure 3. Speedup (left) and parallel efficiency (right) using 128, 256, 512 and 1024 processor cores.

The primary issues that have been demonstrated by these studies are partitioning of the finite element nodes across the processors and communication between the processors. The communication issue is being addressed by continued development of the new BTS method. The partitioning is currently being addressed by implementing a new algorithm that should perform better.

UQ framework and initial applications to fission gas diffusion: Given a computational forward model, with uncertain inputs, our UQ strategy involves a general workflow, relying on probabilistic forward and inverse UQ methods, as follows:

1. Given experimental measurements, use Bayesian inference to estimate a joint density on uncertain model parameters
2. Employ global sensitivity analysis (GSA), coupled with polynomial chaos smoothing and sparsity constraints, to identify the subset of important parameters for UQ
3. Using adaptive sparse quadrature, propagate uncertainty forward through the computational forward model, providing prediction of uncertain model outputs.

Where needed, if the measurement model used in #1 above is computationally expensive, step #3 can be used to build a polynomial chaos surrogate to be used in the Bayesian inference process. Also, and again where needed, we rely on Bayesian model selection to identify the best model for fitting the data in case there are alternatives. Finally, this strategy accommodates coupled models as needed, incorporating the full set of parameters in multiple models as needed.

We are working on employing elements of this workflow as needed on different parts of the project. In particular, we have been interfacing with the domain experts to collect uncertain specifications of reaction parameters computed from density functional theory (DFT) and empirical potential calculations, with associated uncertainties, which will subsequently be used as input for computing species diffusion coefficients at various temperatures using the MARMOT software framework. We have generated samples in the joint space defined by the reported uncertainty in the parameters, and are currently working with the DFT team to compute the diffusion coefficients. These output values will be employed in a Monte Carlo integration approach to compute sensitivity indices (GSA) to determine the fractional contribution of the variance of each input parameter to the variance of each diffusion coefficient.

The results of the GSA effort will be used as a screening heuristic to identify a reduced parameter set to which the diffusion coefficients are sensitive for the purpose of reducing the effective dimension of the uncertain parameter space. This will also be subsequently redone with the full coupled model framework, to identify important parameters in the coupled setting (including Xolotl and MARMOT). The reduced parameter set will be used for the construction of polynomial chaos surrogates for model output quantities of interest (QoIs) employing adaptive sparse quadrature techniques which are particularly useful for high dimensional integration applications. Where experimental data are available, we will use the surrogate models to estimate joint parameter densities on the model input parameters using statistical inference, including assessment of the efficacy of different competing models employing these parameters using model selection in a Bayesian framework. These uncertain parameter estimates will be propagated through the higher-level process models for predicting uncertainties in output QoIs, which ultimately will provide uncertain inputs for BISON.

Experimental Validation targets: Validation of models for the evolution of fission gas bubbles targets experimental databases including a large amount of data for bubble size and density in UO_2 irradiated to various burnups and temperatures. These include that data from Cornell and co-workers [5], Baker [11] and Kashibe et al. [12]. In particular, Cornell et al. [5] examined UO_2 specimens irradiated at a burnup of 3.2×10^{25} fissions/ m^3 and determined the temperature dependence of the intra-granular bubble size and number density. Baker [11] reported measurements from UO_2 specimens irradiated to a burnup of 2.0×10^{26} fissions/ m^3 and different temperatures. Kashibe et al. [12] examined the characteristics of intra-granular fission gas bubbles in irradiated UO_2 fuel pellets before and after post-irradiation annealing, and at different burnups. Also, the more recent thin film ion irradiation studies by He and co-workers [15], in which Kr was implanted into thin films of UO_2 , will provide additional data on implantation rate effects on fission gas bubble evolution.

Finally, an extensive database of fission gas bubble characteristics is available from the experimental study of fission gas behavior in UO_2 conducted by BNFL, UK [16]. The database comprises TEM and SEM examinations of both intra-granular and grain-boundary fission gas bubbles on a large number of specimens from UO_2 fuel specimens that underwent various irradiation conditions, including power ramps and power cycles. The experimental results included in the database can be used to validate models for both intra-granular and grain-boundary bubbles. As for intra-granular bubbles, bimodal bubble populations were observed, which originated from bubble coarsening during the high temperature periods involved in these experiments. Hence, these data provide a means for validation of a bubble coarsening capability, which is under development for the engineering-scale model.

Nuclear Materials Research Activities

The development of materials models for fission gas in UO_2 has made progress on several fronts, directly supporting thrust 1 and 2. We have also worked with thrust 3 to perform a sensitivity study and UQ analysis of a cluster diffusion model developed from DFT calculations and molecular statics. Below we highlight the most important accomplishments in each topical area.

Accelerated Molecular Dynamics and Development of TAMMBER: One of the key goals of the accelerated molecular dynamics (AMD) simulations is to provide fundamental information about atomic scale mechanisms of defects in UO_2 to pass to higher level cluster dynamics and phase field models. However, the astronomical vastness, complexity, and roughness of the potential energy landscape of these defects make the investigation of their long-time dynamical evolution extremely difficult – even for AMD methods – as significant free energy barriers between different regions of configuration space prohibit the

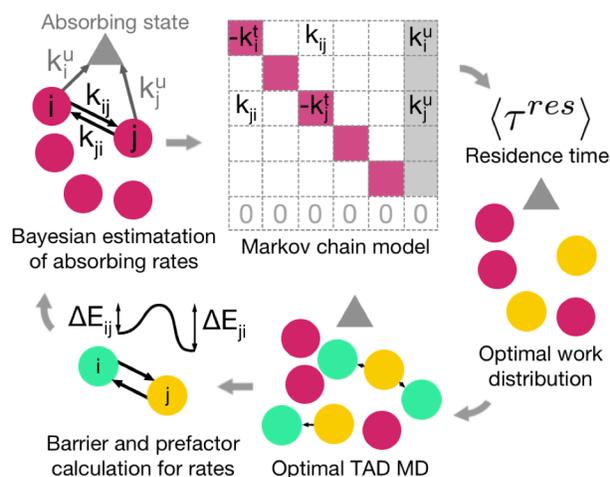


Figure 4. TAMMBER workflow. Using novel analysis of the observed state-to-state transitions, TAMMBER autonomously decides where additional computational work is most profitable to increase the timescales over which the constructed Markov model is valid.

for higher scale simulation schemes such as cluster dynamics. The residence time gives a characteristic timescale over which higher scale simulation trajectories can be trusted; this is a critically important but typically uncalculated parameter in multiscale simulation. With an estimate of the residence time, massively parallel schemes are able to *autonomously* decide where additional computational work will be performed, an essential feature to fully exploit modern supercomputers. The estimators designed in this work are additionally used to optimize where exploration is performed and the degree of temperature acceleration on the fly, giving an autonomous, optimal procedure to explore the state space of complex systems.

TAMMBER has been successfully applied to interstitial defects in bcc iron, routinely reaching residence timescales of hundreds of seconds [17]. However, the energy landscape of point defects in UO_2 feature a large number of low energy barriers that are extremely challenging for any and all exploration methods. Using an isomorphic graph representation of atomistic structures, we are able to significantly compress the energy landscape, as shown in Figure 5. Using the FALCON computer at Idaho National Laboratory, TAMMBER has efficiently harnessed over 2000 cores to produce a kinetic Monte Carlo model of UO_2 point defect clusters that accounts for *all possible* defect transformations over a timescale that currently exceeds tens of milliseconds at 900K; with ongoing computational effort this will soon increase to multiple seconds.

use of direct simulation methods. Indeed, molecular dynamics (MD) simulations of materials are typically restricted to sub-microsecond timescales, a time that is often much too short for a trajectory to cross the barriers that determine the long-time behavior. This makes extrapolation based on short simulations fraught with danger.

We have developed a massively parallel method to build large transition rate matrices from temperature accelerated molecular dynamics trajectories -- **TAMMBER**- Temperature Accelerated Markov Models with Bayesian Estimation of Rates [17]. The essential workflow is illustrated in Figure 4. Novel Bayesian estimators are combined with Markov model analysis to estimate the expected *residence time* in the known state space, providing crucial uncertainty quantification

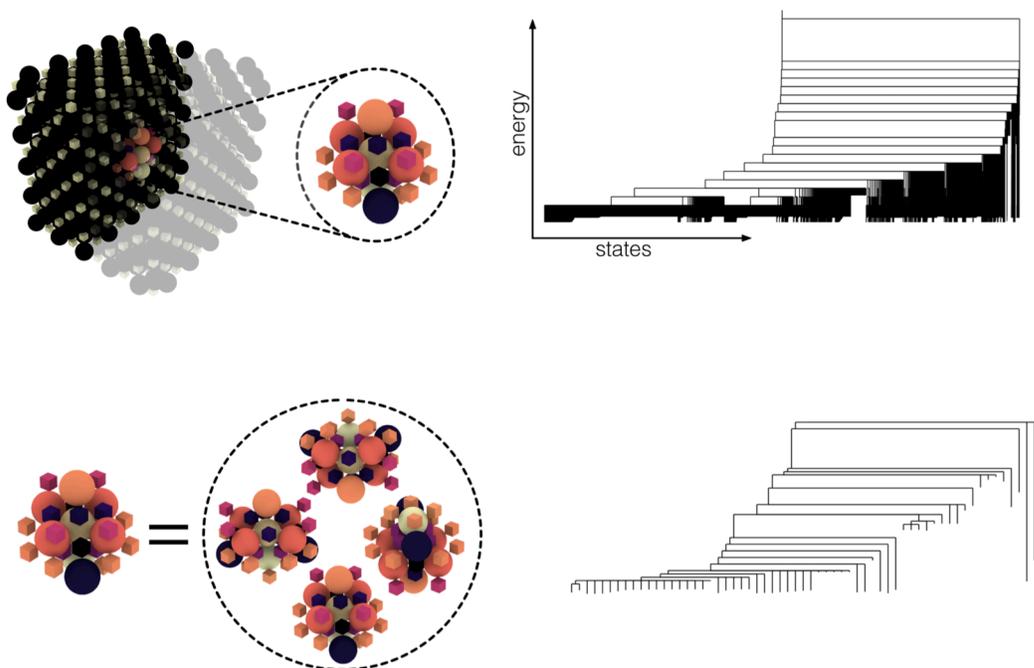
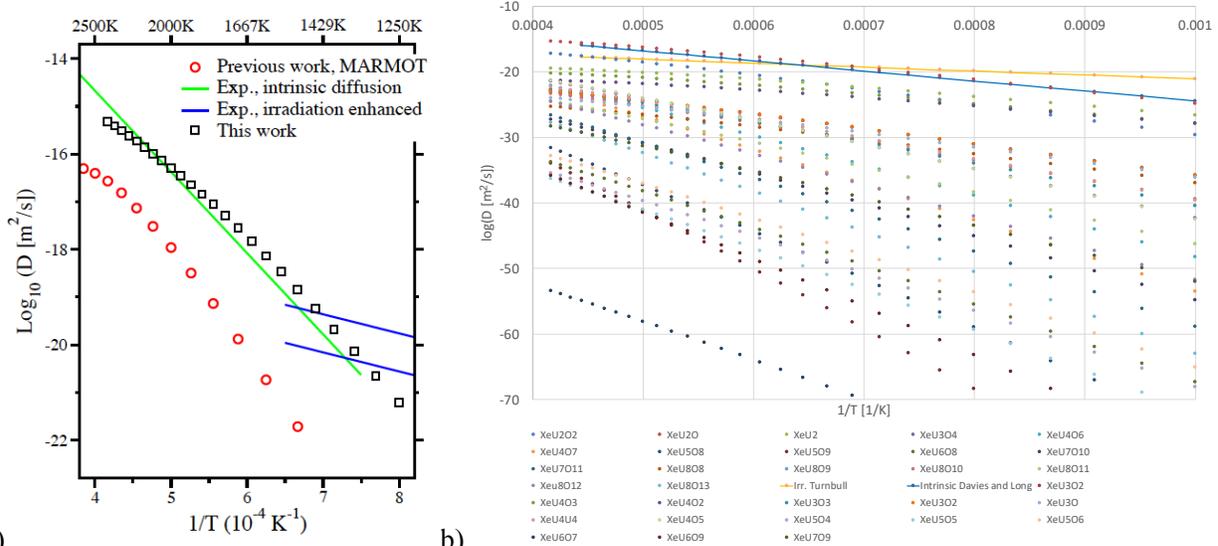


Figure 5. Symmetric simplification of the energy landscape an ‘Anti-Schottky’ point defect cluster in UO_2 . Above left: ‘Anti-Schottky’ point defect cluster. Above right: preliminary energy landscape after one hour of operation on TAMMBER. The presence of multiple low barriers, shown here through a disconnectivity graph, is a major hindrance to exploration. Below left: Graph isomorphism techniques collate many equivalent structures. Below right: The symmetrically compressed landscape allows significant acceleration of the landscape exploration, leading to model validity times of milliseconds and beyond.

Density functional theory and molecular statics calculations of Xe-vacancy clusters: The TAMMBER technique is developed, in part, to validate predictions by DFT and molecular statics calculations of Xe diffusion mechanisms based on clusters consisting of multiple uranium and oxygen vacancies. These calculations have been performed in collaboration with the NEAMS program. The total diffusion rate under thermal equilibrium (intrinsic) conditions as well as that of each individual cluster included in the model are highlighted in Figure 6 [18]. This model shows vastly improved agreement with experimental data compared to previous work [19]. The same data can be used to simulate the behavior under irradiation, which increases the concentration of uranium vacancies and thus the importance of large Xe-vacancy clusters (not shown). The work on radiation-enhanced diffusion due to large Xe-vacancy work is currently being finalized and we are also in the process of subjecting the diffusion model to sensitivity and UQ analysis, as outlined the Computer Science/Applied Math section.



a) The intrinsic diffusion rate of Xe in UO_2 predicted by a model based on DFT and molecular statics calculations [18]. This model shows vastly improved agreement with experiments compared to previous work [19]. b) The diffusion rate of the individual clusters contributing to the total rate in a) (the subscripts denote the number of uranium and oxygen vacancies in the cluster). The XeU_{20} cluster dominates diffusion under intrinsic condition, but the importance of larger clusters increase under irradiation (not shown). Experimental data is shown by solid lines.

Xolotl modeling of intra-granular fission gas bubble evolution: We have evaluated Xolotl-fission code performance against a number of initial test problems to evaluate the implementation of the grouping method developed for larger xenon bubble cluster sizes, and to optimize the code performance (as presented in Figs. 1 and 2). Subsequently, we have begun to benchmark the Xolotl predictions against experimental results on the intra-granular bubble sizes measured by Baker [11]. In these initial calculations, we have not yet incorporated Xe re-resolution and have taken our estimates for Xe gas diffusion from the experimental assessment of Turnbull and co-workers [8]. These experiments involved uranium dioxide fuel pins irradiated in the Atomic Energy Agency (AEA) Winfrith heavy water reactor to burnups of approximately 1% ($\sim 2 \times 10^{26}$ fissions/ m^3) at a power of approximately 8×10^{18} fissions/ $\text{m}^3\text{/s}$ at irradiation temperatures from about 1000 to 1560°C [11]. We have simulated these conditions, to predict the full Xe gas bubble size distribution, as well as the mean size and density.

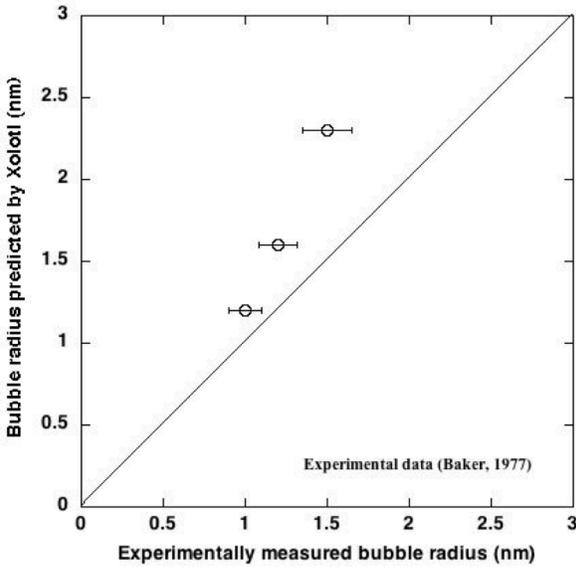


Figure 7. Xolotl-fission predictions of the mean fission gas bubble radius for 3 irradiation temperatures from 1000 to 1560°C at a burnup of approximately 1%, as compared to the experimental data of Baker [11].

Figure 7 compares the predicted Xe gas bubble average size to that measured by Baker [11]. Overall, the agreement in both size and number density (not shown) are quite promising, although the cluster dynamics model predicts larger bubble sizes than experimentally measured. Of course, these initial model predictions do not include a gas re-resolution mechanism, and the current model only incorporates homogeneous gas bubble nucleation whereas the experimental results indicated a strong spatial correlation of bubbles at the lower irradiation temperatures that Baker interpreted as being caused by heterogeneous nucleation along dislocations or fission tracks.

We have also now developed a fission gas re-resolution model based on a systematic evaluation of MD simulations of the interaction of fission product tracks with fission gas bubbles, as a function of bubble size, pressure and interaction geometry with the fission track [20]. The results of this analysis, coupled with a complete assessment of the fission product yields, results in a model for the re-resolution rate of fission gas atoms from bubbles as:

$$b_{het} = 0.25\pi r_c^2 \dot{F} \sum_i y_i \int_{x=0}^{\mu_{c,i}} [1 - e^{-0.05(\zeta S_{e,i} - 9.04)/R^{1.47}}] dx \quad (1),$$

where b_{het} defines the per atom yield of fission gas atoms re-resolutioned into the matrix from a bubble due to the heterogeneous mechanism associated with electronic energy loss. \dot{F} is the volumetric fission density, r_c defines the interaction radius between the bubble and the fission track as the sum of the bubble radius and fission product track radius, y_i is the yield of fission product i , and the sum is over the fission products. The integral is performed over the fission product track length, such that $\mu_{c,i}$ defines the critical range of fission product i at which the electronic stopping power decreases below the threshold required to induce resolution, and the $\zeta S_{e,i}$ represents the effective electronic stopping power of fission product i , and R is the bubble radius [20].

Equation (1) is now being implemented into Xolotl-fission, and future efforts will benchmark the intra-granular bubble evolution model and systematically evaluate the effects of fission gas re-resolution. As well, we anticipate incorporating a more precise input of xenon diffusivity based on lower-length scale modeling, and benchmarking to a larger range of experimental conditions, including higher irradiation temperatures where the intra-granular fission gas bubbles take a strongly bi-modal size distribution.

MARMOT to Xolotl coupling: Our mechanistic model of fission gas release at the mesoscale will capture the fundamental mechanisms of intragranular bubble behavior obtained using the state-of-the-art Xolotl code. It will also describe bubble growth and interconnection on grain boundaries, as well as the formation of fission gas tunnels along grain edges, using the MARMOT code. These two advanced codes will be coupled to fully describe the intra- and intergranular bubbles, as well as the interaction between them. The two codes each move through time, passing information back and forth. Xolotl will calculate the flux of gas atoms to the grain boundaries, passing the fluxes to MARMOT. MARMOT will calculate the nucleation, growth, and interconnection of the bubbles, as well as the grain boundary migration. MARMOT will pass the grain boundary locations back to Xolotl. This coupling is summarized in Figure 8.

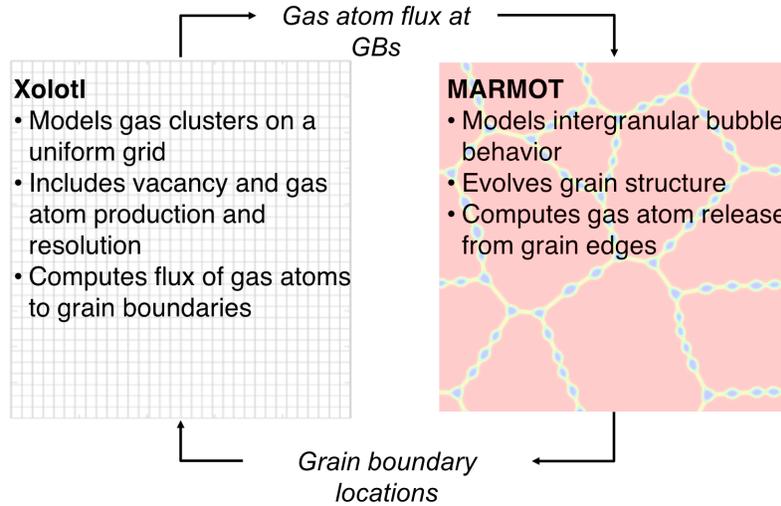


Figure 8. Schematic of the approach that will be used to couple Xolotl to MARMOT for the mesoscale fission gas release model.

Engineering scale modeling: A new reduced-parameter model for the coupled intra-granular bubble evolution and gas atom diffusion to grain boundaries was developed during this initial part of the project. The model is already available in the fuel performance code BISON and is used to efficiently compute intra-granular bubble swelling and the first stage of fission gas release for engineering fuel rod thermo-mechanics calculations.

Additionally, this model will provide a means for the application to engineering fuel analysis of the physical understanding developed through advanced lower-length scale calculations within this project. Such scale bridging cannot be achieved using the empirical models of fission gas bubble evolution currently adopted in engineering scale codes. Further developments of the engineering model will focus on coupling to lower length scale calculations and on model extension to transient behavior and the bubble coarsening process. Basic equations of the current model and a sample of results to date are reported in the following.

Starting from a detailed cluster dynamics formulation, simplifications are applied to obtain a reduced-parameter model, while retaining a physical basis. Defining the total number density of bubbles, N (m^{-3}), the total concentration of gas residing in bubbles, m (m^{-3}), and the average number of atoms per bubble, $n=m/n$, the final form of the model consists of only three differential equations, as follows:

$$\begin{aligned} \frac{\partial N}{\partial t} &= +\nu - \alpha_n N \\ \frac{\partial \psi}{\partial t} &= +2\nu + \beta_n N - \alpha_n \psi \\ \frac{\partial c_1}{\partial t} &= +yF + D\nabla^2 c_1 - 2\nu - \beta_n N + \alpha_n \psi \end{aligned} \quad (2).$$

In Eq. (2), t (s) is the time, y the yield of fission gas atoms, D (m^2s^{-1}) the single atom diffusion coefficient, ν ($\text{m}^{-3}\text{s}^{-1}$) the rate of bubble nucleation, β (s^{-1}) the rate of gas atom trapping at bubbles, α (s^{-1}) the resolution rate, and the parameters α , β are calculated at n .

Initial validation of the new model was performed by comparing results to the experimental data from Baker [11]. A comparison of this model with the experiment are presented in Figure 9 and demonstrate an encouraging accuracy in predicting both bubble number densities and radii. Note that significant uncertainties exist in the parameters, and model improvements will be pursued through coupling with improved parameters (e.g., gas atom diffusivity and resolution rate from bubbles) from the atomistic calculations performed within this project.

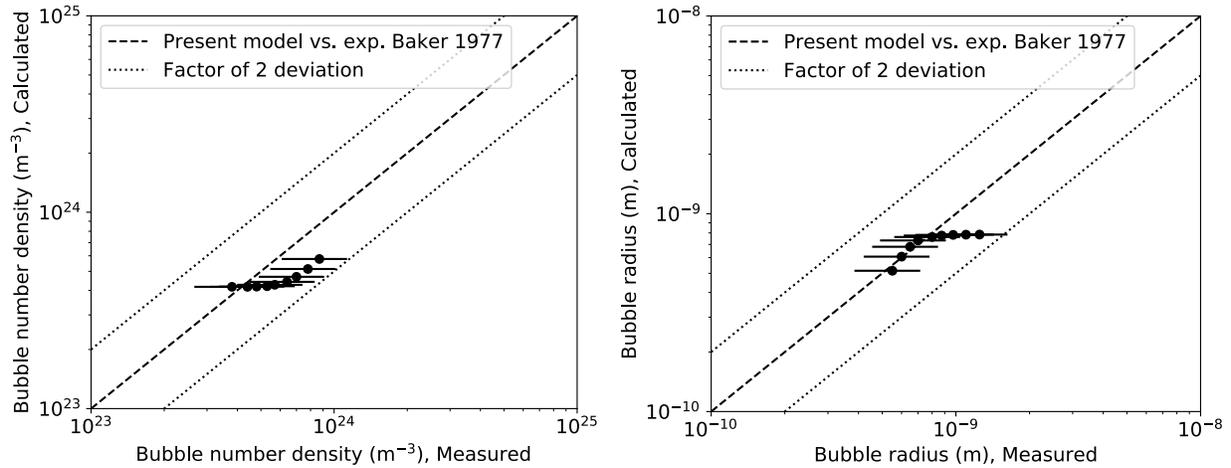


Figure 9. Comparison between the engineering model and the data from Baker [11] for the number density (left) and the radius (right) of intra-granular bubbles in samples irradiated at different temperatures. The horizontal error bars represent the experimental uncertainties.

The model has been implemented in the BISON fuel performance code and applied to simulations of integral fuel rod tests from the Risø-3 experiment [21]. As an example of this activity, Figure 10 compares the BISON calculations to radial distributions of Xe in the fuel grains measured by Electron Probe Microanalysis (EPMA). Such local comparisons are possible for a fuel performance code with a physics-based model of fission gas behavior that includes direct modeling of the fundamental mechanisms. Calculated concentrations of intra-granular fission gas compare very well to the EPMA data. The development of this engineering scale model has led to a journal publication [22].

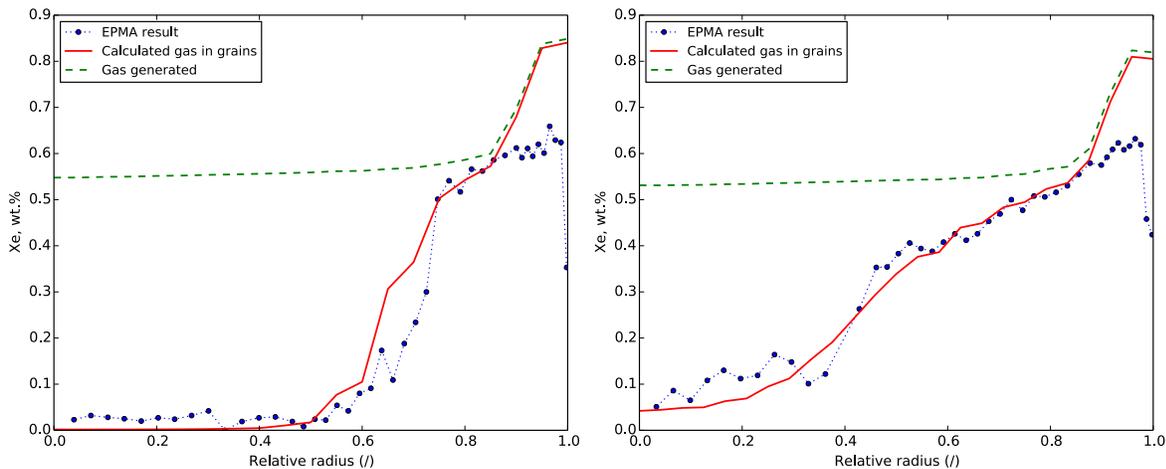


Figure 10. Radial distributions of Xe at the end of life for the Risø-3 AN2 (left) and AN8 (right) fuel rods. BISON calculations are compared to EPMA data. Calculated gas generated is also included.

SUMMARY

This document describes the initial progress of the NE SciDAC project, Simulation of Fission Gas in Uranium Oxide Nuclear Fuel, since the initial kick-off meeting held in December 2017 at the University of Florida. The project has made good progress on the proposed research thrusts and task activities, and will continue to moving forward with its research activities and plans to couple Xolotl-fission and MARMOT. This has been accomplished through the integrated computational and physics research, and the evaluation of different modeling approaches against a series of experimental validation problems that describe gas bubble evolution dynamics. Overall, the project has had a very successful first year, and the project is on track to continue following the research tasks identified in the proposal.

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