UQ framework for fission gas behavior in UO_2 nuclear fuel

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SciDac PI meeting July 22-24, 2018

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Background

- Predictive framework for fission gas behavior involves hierarchies of models across multiple scales, e.g. density functional theory (DFT) calculations (VASP code), molecular statics, cluster dynamics (MARMOT code)
- Input parameters for models at each level are estimated from available experimental data or some high-fidelity models
- 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)

Sensitivity analysis

Identify how model QoIs respond to variation of the model inputs

- Employ a variance based approach, utilizing decomposition of QoI variance invoking the laws of total variance and expectation
- Compute approximations to Sobol indices [1, 2]. The first order indices for each input parameter (S_i) are the fractional contributions of the variation of each parameter alone to the total variance of the QoI. Estimating using Monte Carlo integration:

$$S_i \approx \frac{1}{V}\left(\frac{1}{N}\sum_{k=1}^{N} f(x^k) f(x'^k_{-i} \cup x^k_i) - f_0^2\right) \qquad (1)$$

Statistical parameter inference

When data are available, typically as noisy signals from experiment probes, we can perform Bayesian statistical inference to estimate the parameters of a stochastic model for the data, e.g. for a data model $\mathcal{M}$:

$$z = f(\beta) + \varepsilon \qquad (2)$$

where z is some noisy data, $f(\beta)$ is a model parametrized by a set $\{\beta\}$ for the underlying 'true' data obscured by the noise, and ε is a stochastic element that models the noise (in this case additively). Invoking Bayes' rule:

$$p_{posterior}(\beta|z) = \frac{p_{likelihood}(z|\beta)p_{prior}(\beta)}{p(z)} \qquad (3)$$

our state knowledge of the parameters β given the noisy data z can be expressed as a posterior probability density function (PDF) $p_{posterior}(\beta|z)$, which updates any prior knowledge of the parameters β as represented by a prior PDF, $p_{prior}(\beta)$, with likelihood function relating z to possible instances of β constructed using the data model (eq. 2).

- Compute posterior PDF numerically using Markov chain Monte Carlo (MCMC) methods
- Posterior PDF reveals important correlations between parameters (expected for physics-based models)
- Analysis of $p(z)$ (i.e. implicitly $p(z|\mathcal{M})$) for different choices $\mathcal{M}_i$ (eq. 2) allows for optimal selection of $\mathcal{M}_i$ that is most consistent with the data
- When data is unavailable, have recourse to maximum-entropy (MaxEnt) inference techniques [3]

Polynomial chaos expansions

Efficently represent uncertain input-output mapping using polynomial chaos expansions (PCEs) [4]. For a QoI $f(\beta)$:

$$f(\beta) \approx= \sum_{k=0}^{K-1} c_k \Psi_k(\xi). \qquad (4)$$

an orthogonal polynomial expansion in e.g. Legendre polynomials (Ψ_k) of a uniform random variable ξ

- Coefficients determined using runs of the model
- Coefficients can be estimated using Galerkin projection with quadrature, Bayesian least-squares, Bayesian compressive sensing (regularization)

Extensions

Basis adaptation:

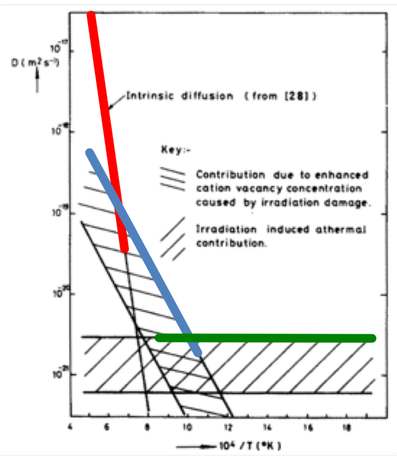
- QoIs are often low-dimensional, variation depends on some combined variation of the input parameters
- Discover a sparse PCE representation using a rotation of the original PCE basis

Multi-level multi-fidelity methods:

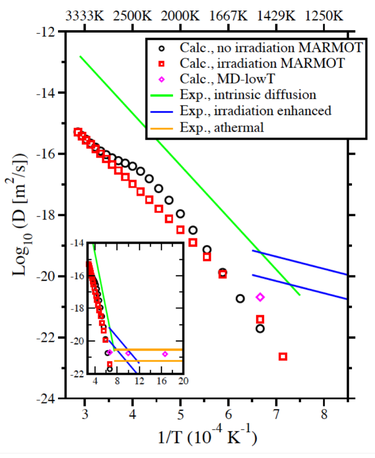
- QoIs can often be estimated using cheaper coarse or lower-dimensional simulations
- When such approximations of the QoIs are available, correlation between high and low fidelity estimates of the QoI can be leveraged to accelerate estimation, e.g. coefficients of PCEs

Density Functional Theory (DFT), molecular statics, and cluster dynamics simulations

- Current empirical models based on experimental data divide the diffusion coefficient for Xe in UO_2 nuclear fuel into three ranges
  - D_1 at high temperature is assumed to be independent of radiation effects (radiation effects are quickly annealed at these temperatures)
  - D_2 at intermediate temperature is increased compared to D_1 as a consequence of radiation damage raising the concentration of vacancies in the material
  - D_3 is caused by direct interaction with the thermal spikes caused by fission of ^{235}U



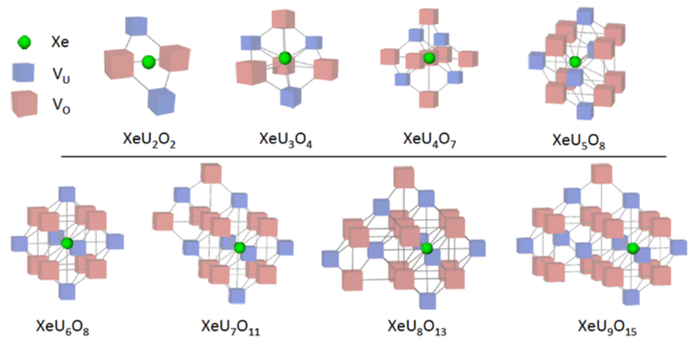
J. A. Turnbull et al., JNM **107,** 168 (1982)



From R. Perriot et al., (2018)

Left: empirical model. Right: simulation using point defect dynamics

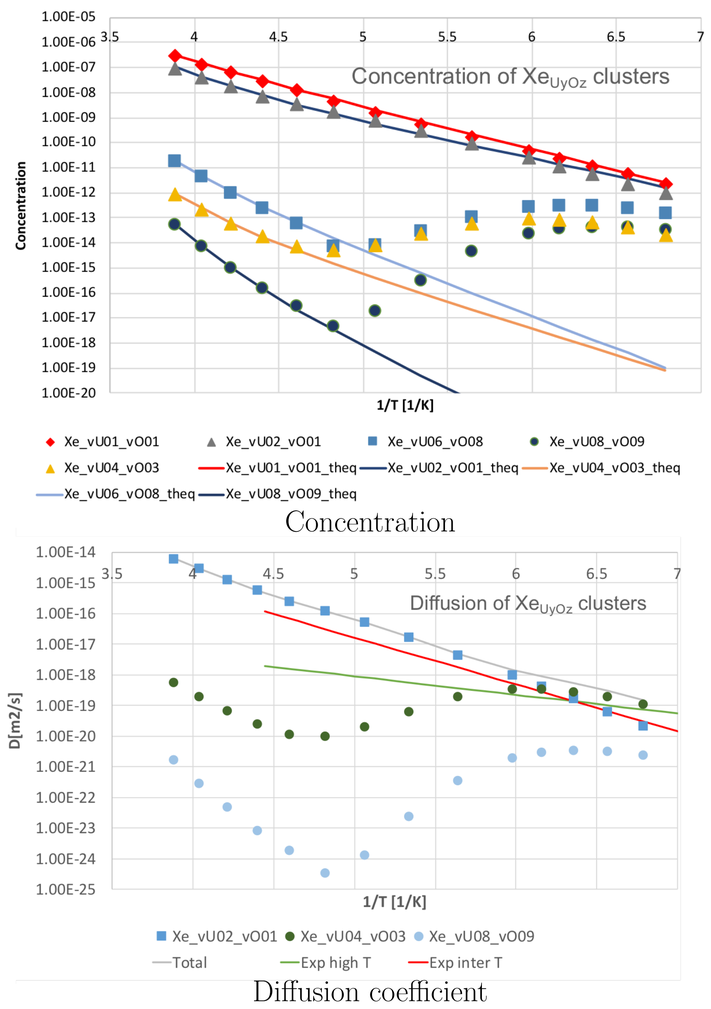
- Existing attempts to calculate D_1 and D_2 in the literature underestimate the diffusion rate compared to the empirical model/experiments.
- Low D_1 and D_2 either due to method issues and/or neglect of important mechanisms, in particular extended clusters under irradiation.
- Calculate D_1 and D_2 fission gas diffusion through simulation of point defect dynamics (small cluster dynamics model in MARMOT) informed by DFT calculations and molecular statics.
- The DFT calculations were performed with the VASP code using a combination of LDA+U (thermodynamic properties of defects) and GGA+U (defect migration barriers).
  - The Hubbard +U term is included to improve the description of the correlated U 5f electrons.
  - GGA+U is used for barriers, because the LDA+U barriers were shown to be too low in relation to experimental data.
  - All defects are modeled as fully charged using the standard approach of adding and removing electrons to the system, including corrections for the mono-pole interaction between defects and shifts of the electrostatic potential.
  - All calculations were performed based on a 3×3×3 supercell expansion of the Fluorite unit cell.
  - The atomic positions, supercell volume and shape were fully relaxed for thermodynamic defect properties.
  - The NEB calculations for migration saddle points were performed at fixed volume using five images.



Xe-vacancy clusters

- Molecular statics calculations were used to estimate defect entropies and attempt frequencies for migration based on the finite displacement method for calculating phonon spectra.
  - The calculations were based on the Cooper-Rushton-Grimes (CRG) potential, which adds a MEAM term to the traditional Buckingham form in order to capture many body interactions.
  - The entropies were calculated with the potential at constant volume and then adjusted for volume change based on DFT calculations.
- The concentration of point defects (uranium vacancies and interstitials) and Xe clusters under irradiation were calculated based on cluster dynamics theory. Oxygen defects were assumed to be in thermal equilibrium at the temperatures of present interest.
- The cluster dynamics model was solved in the MARMOT code, with the defect formation energies and entropies as well as Xe cluster binding energies and entropies obtained the from atomistic simulations.
- The diffusivities can be calculated as an average of the cluster concentrations and their mobility.

Cluster dynamics simulation example

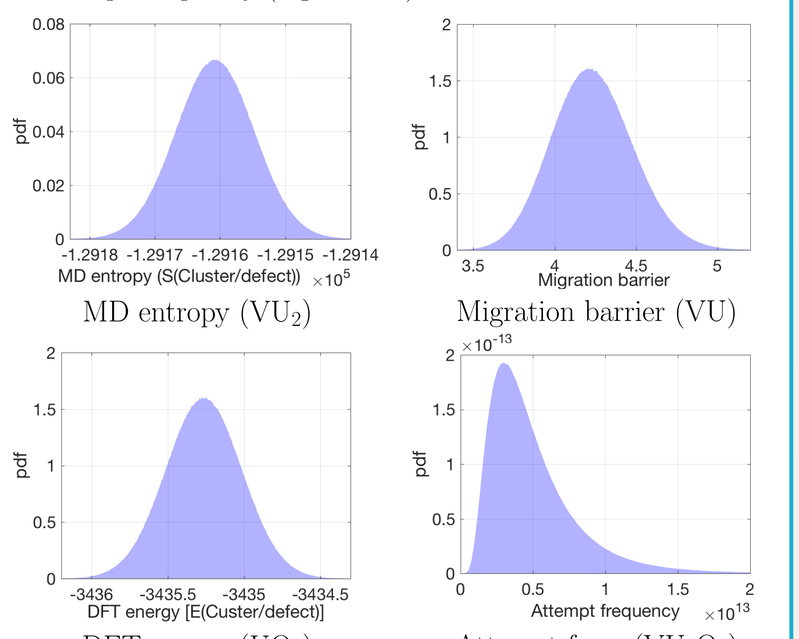


UQ in diffusion coefficient estimation

Initial UQ efforts involve collection of uncertainty assessments of code input parameters. Generally uncertainty is specified as nominal parameters values (means) and standard deviations for unbounded parameters interpreted as Gaussian, and as means and standard deviations of log-normal distributions for parameters that are strictly positive.

Generally four parameters are specified for each of the 46 species:

- DFT energy (Gaussian)
- MD entropy (Gaussian)
- Migration barrier (log-normal)
- Attempt frequency (log-normal)



Input parameter uncertainty structure (PDFs) for select inputs

Computing sensitivity indices in this setting involves drawing samples from these assumed distributions for the evaluation of a multidimensional output, i.e. diffusion coefficients for each species, with Sobol indices computed for each output element separately using eq. 1.

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This research is supported by the U.S. Department of Energy (DOE), Office of Nuclear Energy (NE) and Office of Science (SC), Office of Advanced Scientific Computing Research (ASCR) through the Scientific Discovery through Advanced Computing (SciDAC) program. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525. Los Alamos National Laboratory, an affirmative action equal opportunity employer, is operated by Los Alamos National Security, LLC, for the National Nuclear Security Administration of the U.S. Department of Energy under Contract No. DE-AC52-06NA25396.