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Design and Analysis of Forward and Reverse Models for Predicting Defect Accumulation, Defect Energetics, and Irradiation Conditions

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Motivation and Objective

- Irradiation of materials via electrons, neutrons, and heavy ions cause the generation of point defects (vacancies and self-interstitials) and their clusters of various sizes. The short- and long-term creation, migration, emission, annihilation, and interactions of these defects can lead to detrimental electronic and/or microstructural changes (e.g., void swelling, ductile-to-brittle transition)
- The complexity of microstructural radiation effects makes developing predictive radiation damage models difficult. Current Monte Carlo, rate theory, and cluster dynamics models are quite successful at elucidating damage states and evolution mechanisms.
- However, these models require a **large catalogue of defect formation, binding, and migration energies, and kinetics for all defect species and reactions being considered** to be physically predictive of the resulting damage state.
  - These quantities can be difficult and time consuming to determine.
- A simple "proof of concept" study is performed to identify the extent to which these input and output irradiation parameters (binding and migration energies, dose rate, temperature, defect concentrations and sizes) can be **rapidly determined via regression analysis of large data sets** produced by simple cluster dynamics simulations.

For complete details, see: J.A. Stewart A.A. Kohnert, L. Capolungo, and R. Dingreville Computational Materials Science, **148**, 272–285 (2018).

Cluster Dynamics Modeling

- Spatially dependent balance equation to describe the population evolution of a point defect cluster of size *n* during exposure to radiation:

$$\frac{dC_n}{dt} = D_n \nabla^2 C_n + g_n + R_n(\vec{C}) - D_n k_n^2 C_n,$$

- Various phenomena can be studied with this approach: gas bubbles, dislocation loop nucleation and growth, precipitation, etc.
- $R_n(C)$ represents the net rate of creation, annihilation, and agglomeration of point defects during irradiation conditions.

$$R_n(\vec{C}) = \sum_{i+j\to n} k_{i,j}^{+} C_i C_j - \sum_{i+n\to m} k_{i,n}^{+} C_i C_n + \sum_{m-i\to n} k_{m,i}^{-} C_m - \sum_{n-i\to j} k_{n,i}^{-} C_n + \sum_{m-n\to i} k_{m,n}^{-} C_m$$

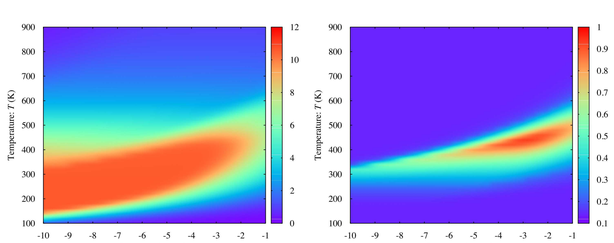

Fig. 1 Average interstitial (left) and vacancy (right) cluster size as a function of dose rate and temperature for pure Fe.

Regression Analysis & Modeling

- Simple linear regression with a quadratic functional dependence is utilized to create surrogate and reduced order models using cluster dynamics results as training data sets.
- A quadratic functional dependence includes linear combinations of individual variables, squares of individual variables, and multiplicative interactions as:

$$Y = \alpha + \sum_{i=1}^{M} (\beta_i x_i + \beta_{ii} x_i^2) + \sum_{i=1}^{M} \sum_{j=i+1}^{M} \beta_{ij} x_i x_j + \varepsilon$$

- Regression models can be developed in various manners, e.g.,
  - **Forward model:** the output quantities from the cluster dynamics model (defect concentration and size) are fit to linear combinations of the input variables (defect energetics and irradiation conditions).
  - **Reverse model:** the cluster dynamics model's input variables (defect energetics and irradiation conditions) are fit to linear combinations of the output quantities concentration and size).
- Perform **global sensitivity analysis** to identify most important quantities describing underlying physics via Sobol' indices:

$$S_i = \frac{\mathrm{Var}_{x_i}[E_{x_{\sim i}}(Y|x_i)]}{\mathrm{Var}(Y)} \qquad T_i = \frac{E_{x_{\sim i}}(\mathrm{Var}_{x_i}(Y|x_{\sim i}))}{\mathrm{Var}(Y)}$$

Forward (Surrogate) Predictions: Damage

- Replace cluster dynamics model with simpler and faster model:
  - Input: defect energetics and irradiation conditions
  - Output: defect concentrations and sizes
- Reduced order model: relevant quantities are temperature, dose rate, and migration energies for vacancy of size 1 and SIA of size 1 and 2

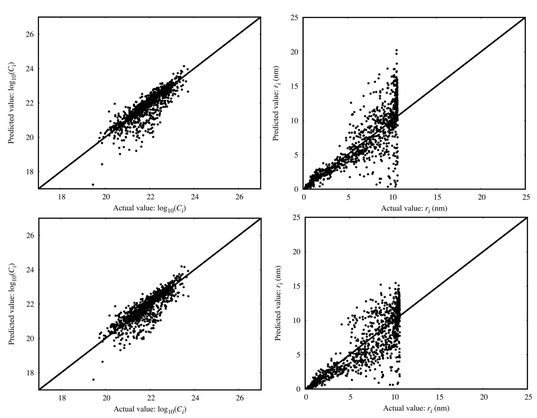

Fig. 2 (Top) Surrogate model predictions and (Bottom) reduced order model predictions for average interstitial defect cluster concentration and size.

Below: Table of mean absolute error and root mean square error quantifying the quality of the simple regression modeling predictions and general reduction in prediction capability for the reduced order models.

	Mean Absolute Error			Root Mean Square Error	
	Surrogate Model	Modified Model		Surrogate Model	Modified Model
C_v	0.12639	0.15907	C_v	0.27737	0.34501
C_i	0.65430	0.74197	C_i	1.49959	1.42516
r_v	0.43559	0.47764	r_v	0.66583	0.69915
r_i	0.25951	0.26965	r_i	0.34412	0.33941

Reverse Engineering Predictions: Energetics

- Determine defect energetics given known experimental information:
  - Input: defect concentrations and sizes, irradiation conditions
  - Output: defect energetics
- Reduced order model: total defect concentration and weighted average of vacancy and SIA defect cluster sizes are used for fitting

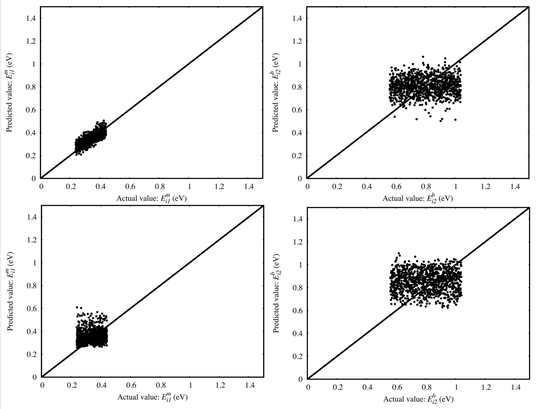

Fig. 3 (Top) Reverse model predictions and (Bottom) reduced order model predictions for the migration energy of a SIA of size 1 and binding energy of SIA cluster of size 2.

Below: Table of mean absolute error and root mean square error quantifying the quality of the simple regression modeling predictions and general reduction in prediction capability for the reduced order models.

	Mean Absolute Error			Root Mean Square Error	
	Reverse Model	Modified Model		Reverse Model	Modified Model
$E_{v_1}^m$	0.08808	0.18426	$E_{v_1}^m$	0.11053	0.25685
$E_{i_2}^m$	0.14170	0.18179	$E_{i_2}^m$	0.17812	0.24457
$E_{v_1}^m$	0.02639	0.02399	$E_{v_1}^m$	0.03748	0.03477
$E_{v_2}^m$	0.15413	0.18086	$E_{v_2}^m$	0.19066	0.22667
$E_{i_2}^b$	0.17329	0.19762	$E_{i_2}^b$	0.21320	0.25177
$E_{v_1}^b$	0.16286	0.18207	$E_{v_1}^b$	0.19825	0.23115
$E_{v_2}^b$	0.18389	0.18892	$E_{v_2}^b$	0.25146	0.25683
$E_{v_3}^b$	0.18641	0.18594	$E_{v_3}^b$	0.26144	0.24484

Forensics Predictions: Irradiation History

- Determine a material's irradiation history:
  - Input: defect concentrations and sizes, defect energetics
  - Output: irradiation conditions (dose rate and temperature)
- Reduced order model: total defect concentration and weighted average of vacancy and SIA defect cluster sizes are used for fitting

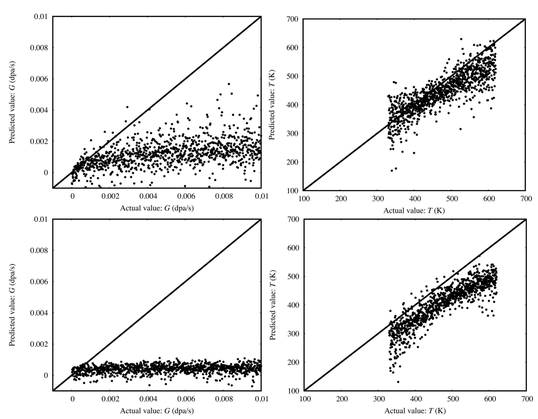

Fig. 4 (Top) Forensics model predictions and (Bottom) reduced order model predictions for irradiation dose rate and environment temperature.

Below: Table of mean absolute error and root mean square error quantifying the quality of the simple regression modeling predictions and general reduction in prediction capability for the reduced order models.

	Mean Absolute Error			Root Mean Square Error	
	Forensics Model	Modified Model		Forensics Model	Modified Model
G	0.87189	0.93247	G	2.79995	1.17509
T	0.08744	0.15301	T	0.11156	0.16987

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