Modeling and Simulation of Primary Damage and Structure Evolution in Ceramics and Metals

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Outline

- Introduction – radiation damage, defects and microstructure evolution
  - Metals
    - Defect production – different structures (bcc, fcc, hcp), temperature.
    - Defects, defect clustering and effects of interatomic potentials
    - Defect production under different conditions – pre-strained crystal, grain boundaries and dislocations
  - Ceramics
    - Ab initio MD simulations of defect generation – threshold energy, charge defects and charge-enhanced migration of defects
    - Defect generation and clustering, defect accumulation and mechanical properties
- Further consideration
  - Kinetic Monte Carlo, phase field modeling and cluster dynamics
Damage Evolution

- damage accumulation (swelling, hardening, creep, ...) requires longer times, investigate with other methods

**Phenomena**

- single displacement cascade
- multiple cascades, cascade overlap
- quenching
- collisional phase
- annealing phase
- point defect/solute diffusion and aggregation
- void swelling, hardening, embrittlement, creep, stress corrosion cracking, ...
- defect and solute migration and clustering
- microstructure evolution
- mechanical property changes

**Methods**

- molecular dynamics
  - ab initio
- kinetic Monte Carlo
  - mean field rate theory
- MD dislocation dynamics
- finite element
  - engineering design
- 3D dislocation dynamics

- APFIM, PAS, TEM, SANS
- dimensional and mechanical measurements

- need to maintain fidelity to physics in spite of approximations necessary for longer time/length scale methods
Defect Production in Different Structures

- bcc – 10 keV, 100 K in Fe

Most of the defects are single defects.

Acknowledgement: The Materials Modelling Group and The University of Liverpool UK
Different sizes and types of clusters are created.
A large vacancy cluster is created (tetrahedral stacking fault)

Defect Production in Different Structures

fcc – 25 keV, 100 K in Cu

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Fig. 1. Log-log plots of $N_F$ vs $E_p$ for Ni$_3$Al and the pure metals Cu, Fe, Ti and Zr at 100 K, and Al and Ni at 10 K, demonstrating the power-law dependence of Eq. (2) in the text. The inset table shows the values of $m$ and $A$ (with $E_p$ in keV) obtained with the best-fit lines shown in the figure. The data for Al and Ni were kindly supplied by Almazouzi et al. [20].

Fig. 3. The fraction, $f_i^{cl}$, of SIAs that survive in clusters of two or more in Cu, $\alpha$-Fe, $\alpha$-Ti, $\alpha$-Zr and Ni$_3$Al at 100 K.

Bacon, et al, JNM 276, 2000

- different values, but similar power-law depending in defect survival
- energy dependence of SIA clustering similar, varies by a factor $<2$
Defects and Defect Clustering in bcc, fcc and hcp

- Different Fe interatomic potentials

- No large difference in the number of FPs, but slightly difference at high PKA energy.
- Slightly different clustering – both interstitials and vacancies.
- The scatter of prediction by recent developed potential is smaller than the previous.

Recent work on Cu (X. M. Bai et al. – Science 327 (2910) 1631)

- GBs – a surprising “loading-unloading effect.
- Interstitial can load into the GB, which acts as a source, emitting interstitials to annihilate vacancies in the bulk.
- This is efficient for annihilating immobile vacancies in the nearby bulk.
Pre-strain Effect on Cascades in Zr

- Zirconium Strain Effects

Local strain in zirconium near hydride could be very high (up to 6%)

A large number of voids are formed at the interface under irradiation

Pre-strain Effect on Cascades in Zr

- Strain Effects (2-D video)
  - 5% uni-axial strain along c-axial, 10 keV

- Void formation at the core of damage region, along with the emission of dislocations
- Dislocations transport atoms from damage region
- Dislocation accelerate void formation

Cascade Interaction with Dislocation

- Formation of stacking fault tetrahedron near the dislocation line.

Primary Damage in Ceramics

- Large-scale *ab initio* MD simulation of defect creation $\sim E_d$
  - SIESTA and NWCHEM codes
  - SIESTA - basis set: linear combination of numerical atomic orbitals (LCAO)
  - NWCHEM – basis set: plane wave
  - Ion-electron interaction: normconserving Troullier-Martins pseudopotential
  - Number of atoms – 128 or 1000 atoms.

- We have further developed the SIESTA code to include:
  - Initiate PKA at a given energy and a recoil direction
  - Conservation of energy and momentum
  - Variable time steps ($1.0 \text{ fs} \sim 0.001 \text{ fs}$), which is based on energy and displacement criteria

- SiC, $Y_2Ti_2O_7$, GaN, zirconate pyrochlores ($Nd_2Zr_2O_7$, $Sm_2Zr_2O_7$, $La_2Zr_2O_7$), $Gd_2Zr_2O_7$, $Gd_2Ti_2O_7$, ThO$_2$, CeO$_2$, ZrO$_2$ and SrTiO$_3$
Primary Damage in Ceramics

- C [-1-1-1] Recoil

- Strong back recoil – leading to the formation of a C-C split interstitial

45 eV

47 eV

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Primary Damage in Ceramics

C [-1-1-1] Recoil

Note that the charge state in (B) is different from that in (D) - $\Delta q$
Primary Damage in Ceramics

- a C recoil – 47.5 eV

- A charge-driven formation of defects – important for covalent and ionic materials
- Some charges accumulate on the C vacancy
- Accumulated charge is not in the center of the vacancy, and the strong interaction of the charged vacancy with neighbors leads to a significant Jahn-Teller distortion
- C vacancy is in a positively charged state (fractional charges)
- All the results are consistent with previous experimental observations
- Present simulations show a dynamic process of how a charged defect is formed

Charge-density contour plot for the final atomic configuration (E) in a (1-10) plane

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Primary Damage in Ceramics

- MD simulation of defect accumulation in SiC
  - The cascade overlap is simulated in a block of $10a_0 \times 10a_0 \times 50a_0$.
  - The Si PKA energy is 10 keV, and temperature is 200 K.
  - Each cascade is ended at 10 ps, following by another 10 ps to control temperature.
  - Total 140 cascades are simulated, and dose rate is about $10^8$ dpa/s.
Primary Damage in Ceramics

- Cascade overlap and defect accumulation

- Both interstitials and antisites are accumulated during overlap process.
- Small clusters nucleate at low dose and grow into larger clusters.
- The completely disordered state (0.28 dpa) – homogenous distribution of defects.
Comparison with Experiments

- **Relative disorder**

  ➔ Both C and Si relative disorder increase with increasing dose, showing a sigmoidal dependence on dose.

  ➔ In general, the simulation results are in good agreement with experimental observations.

  ➔ The increase in relative disorder at low dose is due to single defect accumulation and small cluster nucleation.

  ➔ The formation of amorphous domains (clusters) contribute to the rapid increase in the relative disorder at higher dose.

Comparison with Experiments

Experimental HRTEM Images

HRTEM Image Simulation of Overlapped MD Cascades
Volume Change From MD Cascade Overlap

Predicted Saturation Swelling is in reasonable agreement with experimental value

Experimentally, Saturation Swelling = 10.8% (Snead and Hay, 1999)

Mechanical Properties due to Cascade Overlap

- Elastic constants, elastic modulus and bulk modulus decreases with increasing dose.
- The reduction in these values is fast at low doses, but slow at high doses.
- The most reduction is due to accumulation of point defects and small clusters.
- The contribution due to topological disorder associated with amorphous domains is small.
Mesoscale Simulation of Defect Evolution

- Kinetic Monte Carlo Approach

- The total numbers of point defects decreases with increasing temperature.
Mesoscale Simulation of Defect Evolution

- Phase Field Model of Gas Bubble Growth in UO$_2$

Simulation: Temporal evolution of gas bubbles (the color denotes gas concentration)

The model takes into account:
- Diffusion of He atoms and He-V clusters
- Structural defects
- Elastic interaction
- Inhomogeneity of diffusivity
- Inhomogeneity of elasticity

The model can predict:
- Gas bubble evolution
- Gas release
- Thermal conductivity evolution
- Mechanical property evolution

Experiment: Gas bubbles
Mesoscale Simulation of Defect Evolution

Cluster Dynamics Simulation of Void Swelling in Fe-Cr Alloy

Effect of Cascade Vacancy Clustering
SUMMARY

- Multi-scale approach represents a great tool for studying materials behavior under various conditions.
- *Ab initio* MD can be used to address charge transfer and electron excitation.
- Computer simulations are useful to design new radiation tolerance materials.
- The methods to simulating long-time defect evolution need to be further explored.