Wednesday, January 30, 2008

Multiscale Modeling of Defect Mechanics in Materials for Advanced Nuclear Energy

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10-11am, Building 5700, RM D307
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ABSTRACT

The key to developing advanced materials for nuclear applications is in understanding the interplay between the various physical scales present, from atomic level interactions, to microstructural composition and macroscale behavior. Defect mechanisms at the atomic scale determine the microstructure and ultimately, a whole host of macroscopic properties, including radiation resistance, strength, ductility, toughness, thermal conductivity etc. This talk focuses on the mechanics and kinetic properties of point defects (vacancies, interstitials, external atoms, voids) and line defects (dislocations) in materials used for nuclear applications. Computational algorithms and techniques including Monte Carlo, molecular dynamics, linear elasticity and electronic structure methods are used to connect nanoscale defect mechanisms to their macroscopic manifestations. I examine how defects behave in nuclear fuel and structures under ambient and radiation damage environments, such as those present in a nuclear reactor. Specific cases include the behavior of oxygen interstitials in uranium dioxide and radiation damage in iron. Hierarchical approaches presented here have wide applicability in connecting nanoscale mechanisms to continuum-level observables especially when structural behavior can be parsed into several scales, each with its own distinct characteristics.

Multiscale Materials Modeling and Nuclear Engineering

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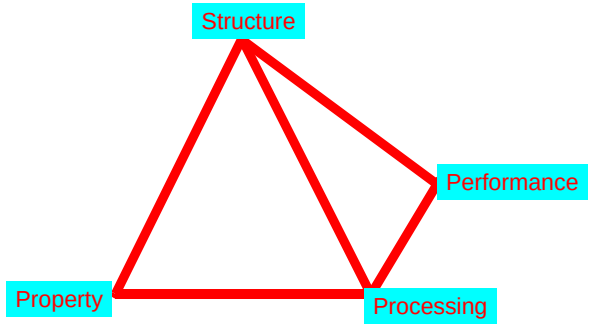
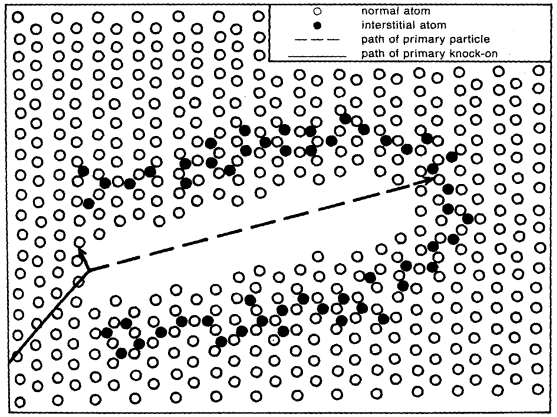
Materials Science for Nuclear Reactors

- Radiation Effects in Materials
- New Materials and Properties
- Nuclear Materials Technology

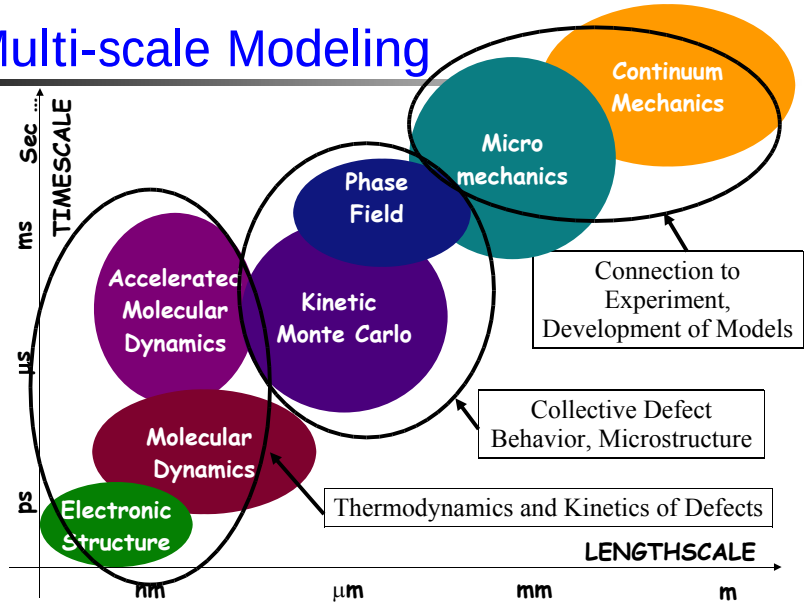


Materials Modeling: Radiation Damage

- George Vineyard was the first to study the dynamics of radiation damage using computer simulation Phys. Rev. 120, 1229 - 1253 (1960)

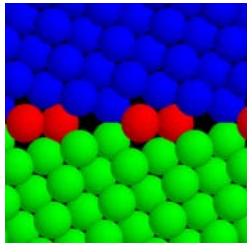


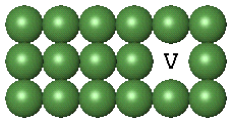
Multi-scale Modeling

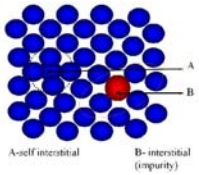


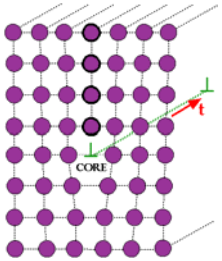
Defect Mechanisms in Materials

- Defects, even in very small concentrations, can have a dramatic impact on the properties of a material
- Without defects:
  - Engineering materials would be much stronger
  - Solid-state electronic devices would not exist
  - Metals would not be formed, rolled or extruded
  - Ceramics would be much tougher
  - Crystals would have no color
- Strength, Ductility, Fracture, Corrosion, Friction ….
- Resistivity, Current density, Luminescence ……
- **Varied dimensionality:** point, line, planar, volumetric





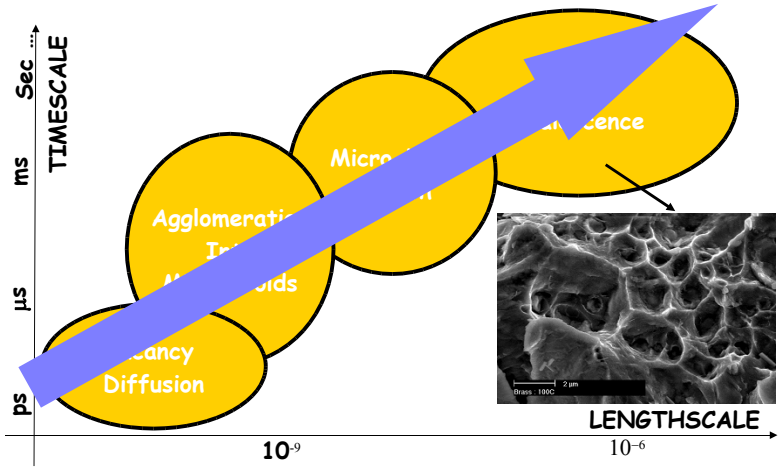




Defect Mechanisms in Nuclear Materials

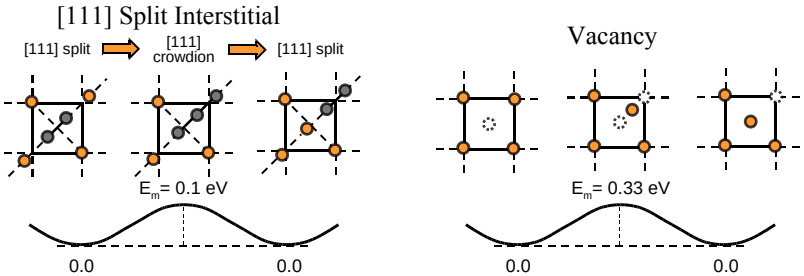
e.g., Radiation Induced Swelling and Fracture

Multiple Length and Time Scales Involved !!



Electronic Structure Calculations

- Density Functional Theory (DFT)
  - Treat electrons quantum mechanically, Treat ions classically
  - No fitting parameters
  - Results:
    - Defect energetics, formation energies, structure
    - Phonon spectra
      - T-dependent energetics of defects



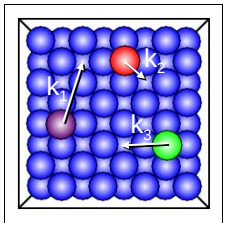
Molecular Dynamics

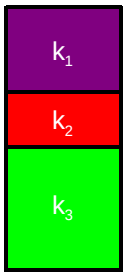
- Each atom is treated as a point mass
- Simple force rules describe the interactions between atoms
- Newton's equations are integrated to advance the atomic positions & velocities

$$F_i = m_i a_i = -\nabla_i V \qquad -\frac{dV}{dr} = m_i \frac{dr_i^2}{dt^2}$$

- Statistics are extracted from the motion of the atoms to get thermodynamic and mechanical properties
- Semi-empirical potentials fit to ab-initio and experimental data
  - Cohesive energy, lattice constant (a_0) , elastic constants (c_{11}, c_{12}, c_{44})
  - vacancy formation energy, stacking fault energy

Kinetic Monte Carlo Algorithm





- Have system of rates k_i
- E.g. for diffusion and transport, k_i's are migration rates
- Select rate at random weighted by probability

$$\frac{\sum_{i=1}^{m-1} r_i}{\sum_{i=1}^{M} r_i} < \zeta_1 < \frac{\sum_{i=1}^{m} r_i}{\sum_{i=1}^{M} r_i}$$

- Advance clock

$$\delta t = \frac{-\ln(\zeta_2)}{\sum_{i=1}^{M} r_i}$$

- Requires knowing rates k_i
- Rates supplied by atomistic simulations

Continuum calculations

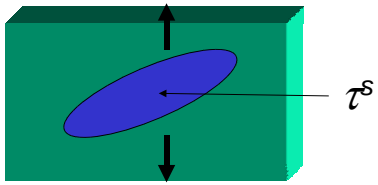
E.g. Visco-Plastic Self Consistent Model

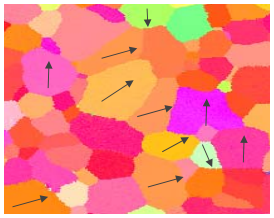
- Relates individual grain deformation to polycrystal behavior
- Grain deformation depends on shape, crystallographic orientation, interaction with medium
- Visco-plastic law assumes threshold shear stress τ_s for slip

$$\dot{\varepsilon}_{ij} = \dot{\gamma}_o \sum_s m_{ij}^s \left(\frac{m^s : \sigma}{\tau^s} \right)^n$$

- Threshold stress τ_s is strongly related to microstructure
- The aggregate is represented by a collection of crystal orientations with volume fractions chosen to reproduce the texture.

12 slip systems {111} <110>





Lebensohn and Tome, *Acta Metall Mater*, 1993, **41**, 2611-24

UO_2 Fuel

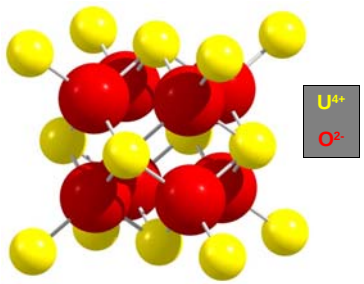
- The most commonly used nuclear fuel

Pros

- Fluorite structure
  - Radiation tolerant
  - Can accommodate fission products with little swelling
  - Can accommodate other actinides such as Pu, Th, etc. without changing the structure
- High melting point (~3050 K)
- Chemically stable

Cons

- Low thermal conductivity
- Higher cost of processing compared to metals



Rigid-ion Models for UO_2

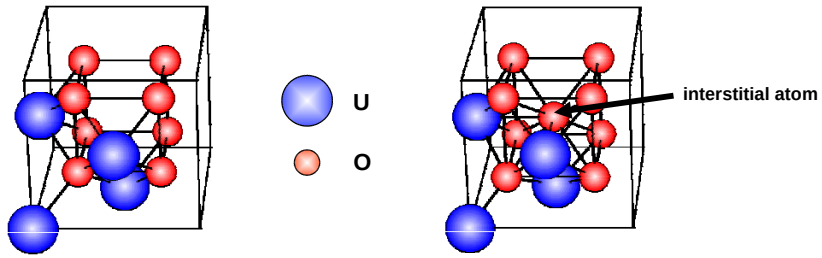
Besides the long-range electrostatic interactions:

- Grimes' model
  - Buckingham potential
  - Parameters given by Grimes et al.
  - No short range U-U interaction
  - Integral charges (z_U=+4,z_O=-2)
  - Highly transferable (U^{5+} and other fission products can be included)
- Yamada's model
  - Includes Morse term (represents "covalence")
  - Parameters given by Yamada et al.
  - Fractional charges (z_U=+2.4,z_O=-1.2)

$$V(r) = A\exp\left(-\frac{r}{\rho}\right) - \frac{C}{r^6}$$

$$V(r) = A\exp\left(-\frac{r}{\rho}\right) - \frac{C}{r^6} + D\left[\exp\{-2\beta(r-r')\} - 2\exp\{-\beta(r-r')\}\right]$$

Defect Mobility in UO_{2+x}



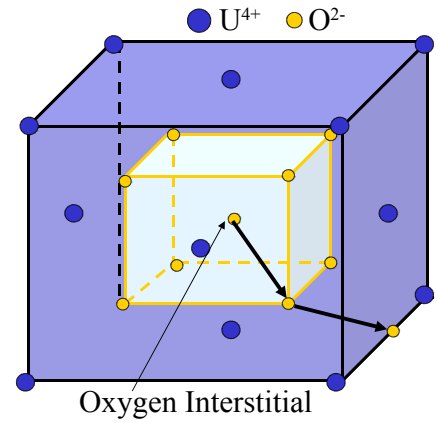
Structure of UO_2 Structure of O interstitial

Defect	Energy (eV)
V_O^{++} diffusion	0.52
O_i^{--} diffusion	1.41
O_i^{--} binding to U^{5+}	0.63
O_i^{--} binding to (O_i^{--} + U^{5+})	> 0.31 (in progress)

Next: feed into higher level models

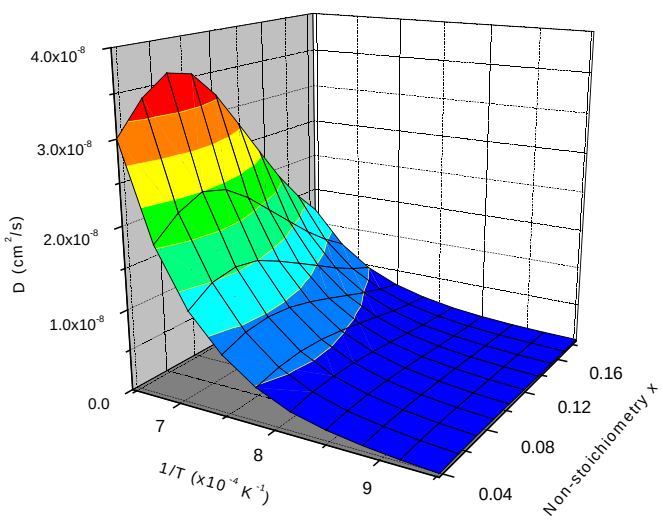
KMC Simulation of Oxygen Diffusivity in UO_{2+x}

- Interstitialcy mechanism of migration
- Blocking model employed to describe the interactions between two oxygen interstitials
- Calculate the mean square displacement of the oxygen interstitials



$$mean\,square\,displacement = \langle \Delta r^2 \rangle = 6Dt$$

Oxygen Diffusivity in UO_{2+x}

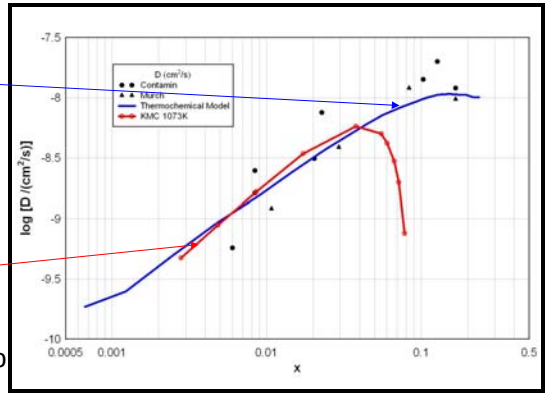


Oxygen Diffusivity in UO_{2+x}

- Thermo-chemical model for oxygen diffusivity (fit to experiment)

$$D_s = D_0 x \exp(-\frac{E_0}{k_B T})\exp(-\Theta x)$$

- Diffusivity obtained by KMC has the same form and magnitude as experiment and thermo-chemical model
- KMC model turns over too fast
  - Need collective behavior of interstitials

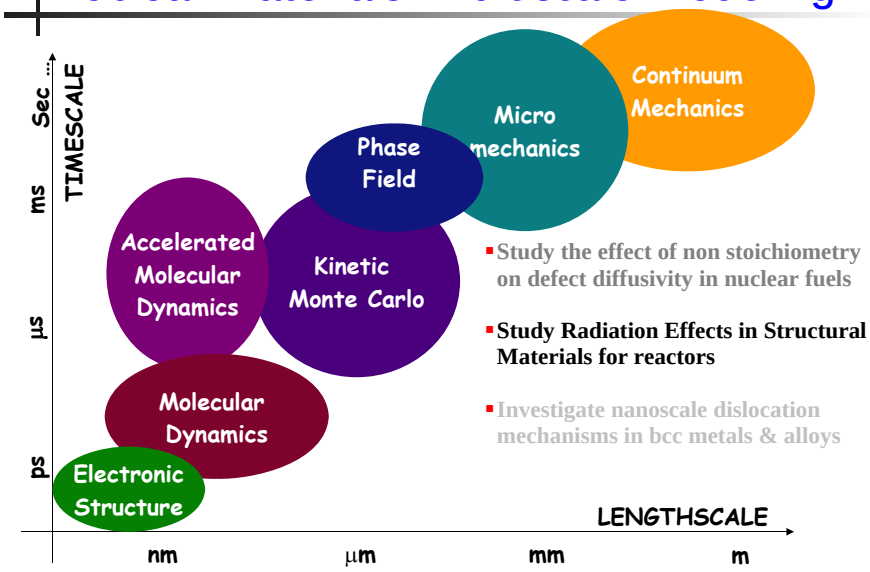


No fitting parameters in MD-KMC results except for interatomic potential

Conclusions: Oxygen Diffusivity in UO_{2+x}

- Thermo-chemical model for oxygen diffusivity (fit to experiment) and KMC model are in agreement at low non-stoichiometry values
- Diffusivity obtained by KMC has the same form and magnitude as experiment and thermo-chemical model
- KMC model turns over too fast at higher stoichiometric values
  - Need collective behavior of interstitials
  - "Willis" clusters formation and mobility
  - Grain boundary diffusivity at higher non-stoichiometric values

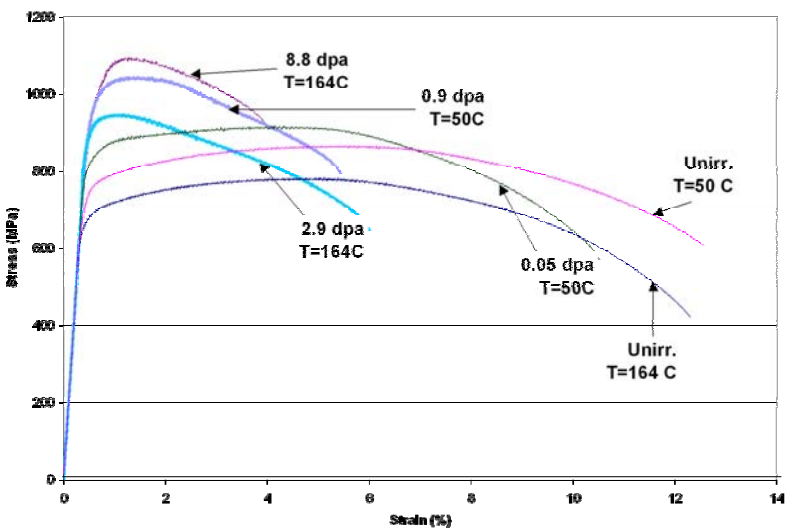
Nuclear Materials: Multiscale modeling



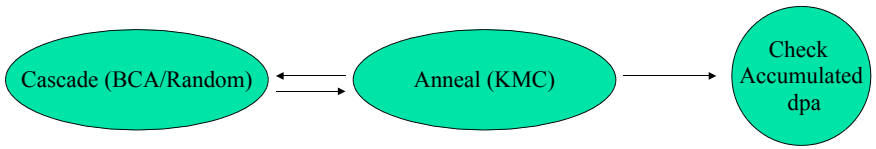
- Study the effect of non stoichiometry on defect diffusivity in nuclear fuels
- **Study Radiation Effects in Structural Materials for reactors**
- Investigate nanoscale dislocation mechanisms in bcc metals & alloys

High Energy Irradiation

Mod 9Cr-1Mo



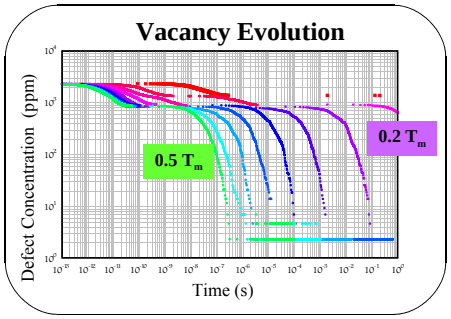
Continuous Damage KMC

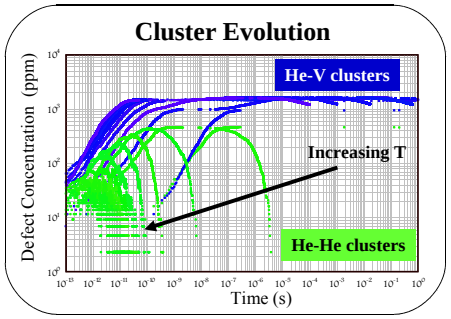


- Cascade debris introduced at periodic intervals
- System annealed using KMC simulations
- KMC simulations
  - Migration of gas, vacancies, SIA, clusters and bubbles
- Variables
  - Incident Energy (eV) – size of cascade
  - He – displacement ratios (appm/dpa)
  - Damage (dpa)
- Compute bubble density and gas concentration in bubbles

Temperature Effects on Defect Evolution

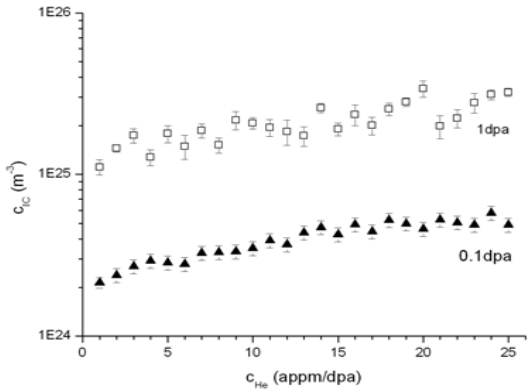
- **Decrease in Vacancy concentration**
  - Helium migration to vacancies
  - Vacancy migration and coalescence
- **Cluster Lifetimes**
  - Helium-Vacancy clusters persist over longer times than He-He clusters





Interstitial Cluster Density

- Interstitial Cluster density shows weak dependence on helium concentration, strong dependence on dose (dpa)
- TEM observation suggest no correlation between helium content and interstitial cluster density (Sencer 2003, Maloy 2001)



Connect to Viscoplastic Model

- The critical stress for moving dislocation contains two contributions: a forest hardening term and an irradiation hardening term

$$\tau = \tau^{forest} + \tau^{irrad}$$

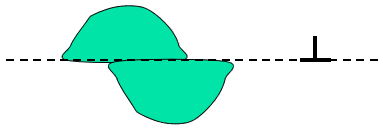
- The forest hardening is a function of the shear 'Γ' accumulated in each grain

$$\tau^{forest} = \tau_0 + (\tau_1 + \theta_1 \Gamma)\left\{1 - \exp\left(-\frac{\Gamma\theta_0}{\tau_1}\right)\right\}$$

- The irradiation hardening term is a function of the evolving defect size 'd', the defect density 'N' and the strength of the dislocation-defect interaction 'α'

$$\tau^{irrad} = \alpha\mu b\sqrt{N.d}$$

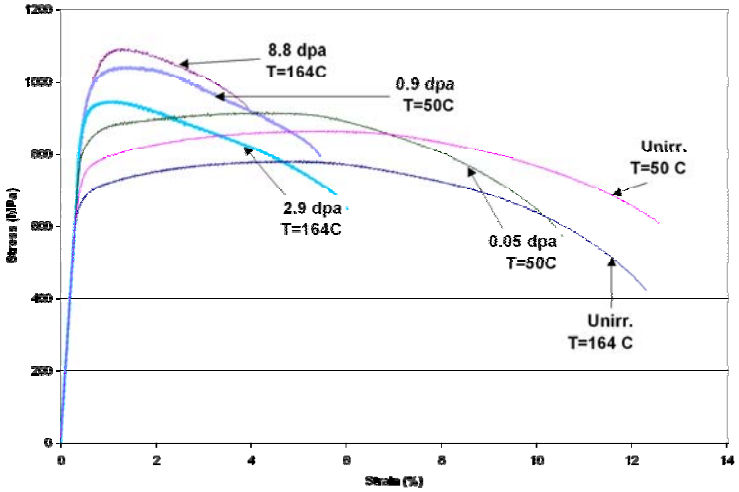
- Obtain *d* and *N* from KMC, α from MD



Lebensohn et al., *Acta Metall Mater 2003*, Arsenlis et. al, *Phil Mag, 2004*

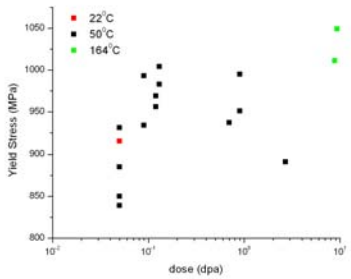
High Energy Irradiation

Mod 9Cr-1Mo



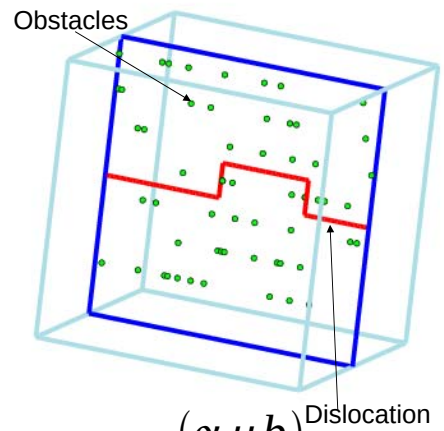
Maloy et al. Journal of Nuclear Materials 2001

Irradiation effects on Yield Stress



Maloy et al. Journal of Nuclear Materials 2001

Dislocation Defect Cluster Interactions



$$\Delta\sigma \propto \frac{(\alpha\mu b)}{L}$$

- Defect Clusters act as obstacles to dislocation motion
- More stress is needed to move dislocation
- Effect is hardening of material
- MD/KMC calculations give number density and size of defects
- Introduce dislocation-defect interaction to viscoplastic code

Radiation Damaged Microstructures

- **Diffraction:** e.g. x-ray, neutron diffraction for defect geometry
- **Spectroscopies:** e.g., NMR, PALS, THDS for defect energies, concentration
- ***In-situ* TEM:** TEM interfaced with ion accelerators for microstructural evolution

Atomistic methods
- First principles calculations
- Semi-Empirical (EAM,MEAM) potentials
- Accelerated dynamics methods

Kinetic Monte Carlo
- Stochastic event based simulation
- Parameterised by atomistic calculations and experiments

Microstructural Viscoplasticity Model
- Model for rate-dependent plasticity
- Stress-strain response of polycrystal
- Input from MD/KMC:
  - Bubble density, size
  - **Dislocation-defect interaction**

Mechanical Testing - 3 pt. bend, tensile, shear punch and compression to obtain stress-strain curves

Nuclear Materials: Multiscale modeling

- **Global Nuclear Energy Program**
  - Nuclear Engineering – Materials issues are at the forefront
  - Fuel and structural properties depend on nanoscale defect mechanisms
  - Combining multiscale computational materials science techniques with experimental methods key to understanding materials issues
- **Hyperstoichiometric UO_{2+x}**
  - Oxygen interstitial diffusivity calculated using multiscale methods
  - Non-Stoichiometry dependence of oxygen interstitial diffusivity investigated
- **Radiation effects in structural materials**
  - Helium evolution in structural materials investigated using molecular dynamics, kinetic Monte Carlo methods
  - Defect density and size connected to polycrystalline model to calculation yield stress dependence