

# Reactor Materials Modeling: A Multiscale Challenge

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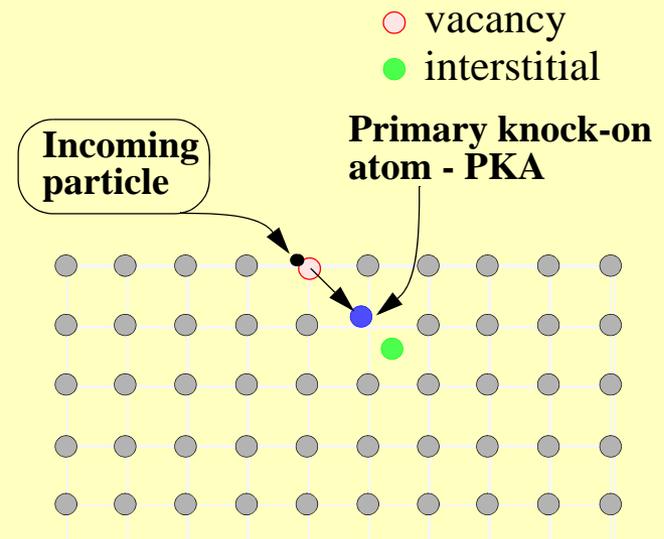
# Why do we care about radiation damage in structural materials - What is the impact?

Desirable material properties: strength, ductility, toughness, dimensional stability, are all largely determined by the nature of their defect structure

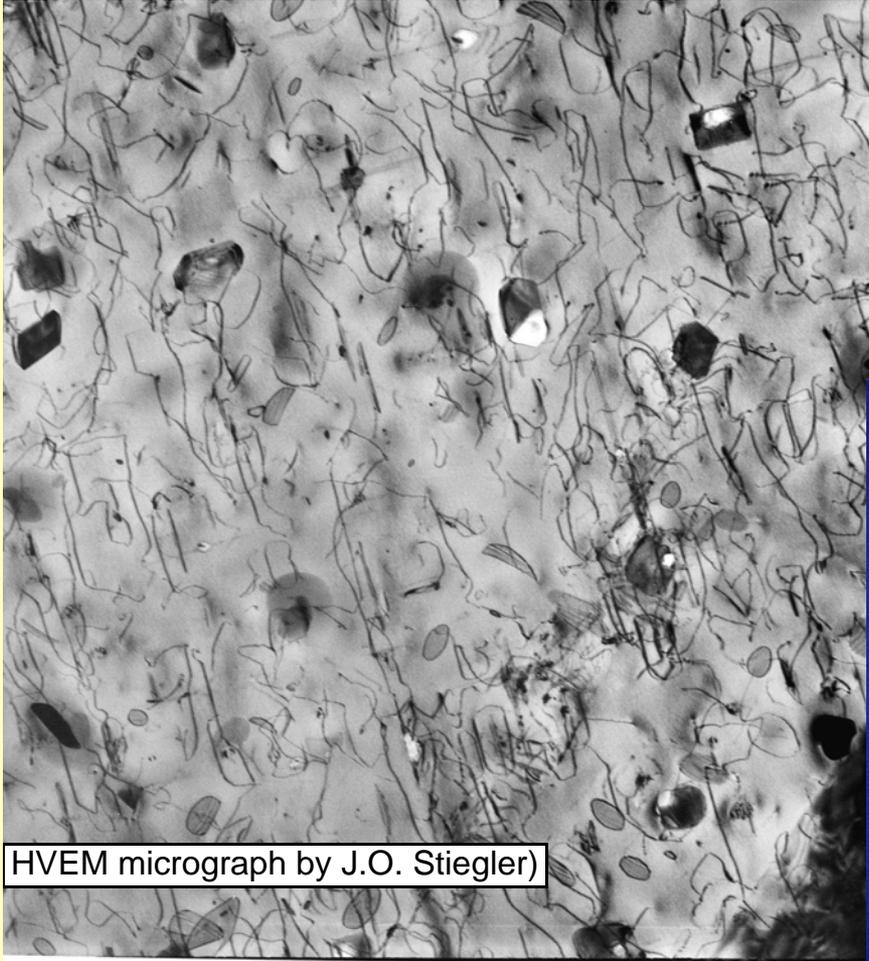
- grain size, other internal interfaces
- dislocation density
- size and density of second phase precipitates

Irradiation with energetic particles leads to atomic displacements

- neutron exposure can be expressed in terms of particle fluence ( $\#/m^2$ ) or a dose unit that accounts for atomic displacements per atom - **dpa** (standard by Norgett, Robinson and Torrens, see ASTM E693)
- lifetime component exposures are in the range of  $\sim 0.01$  to more than 100 dpa
- cumulative impact of atomic displacements: radiation-induced evolution of pre-existing microstructure and the formation of new defect structure



## Example: Radiation-induced microstructure in austenitic stainless steel



- Frank faulted dislocation loops, dislocation network
- additional phases, some non-equilibrium
- large voids

### IRRADIATION - INDUCED SWELLING

20% CW 316  
823°C

1 cm

UNIRRADIATED  
CONTROL

$1.8 \times 10^{23} \text{ n/cm}^2$   
(E > 0.1 MeV)

HEOL 8802-010

# Why do we care about modeling radiation damage in structural materials?

Although irradiation experiments can not be replaced by modeling alone, a purely experimental approach to understanding the effects of irradiation is also not practicable

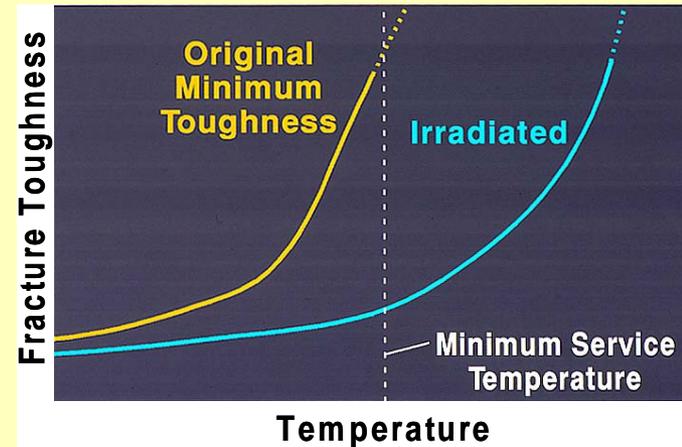
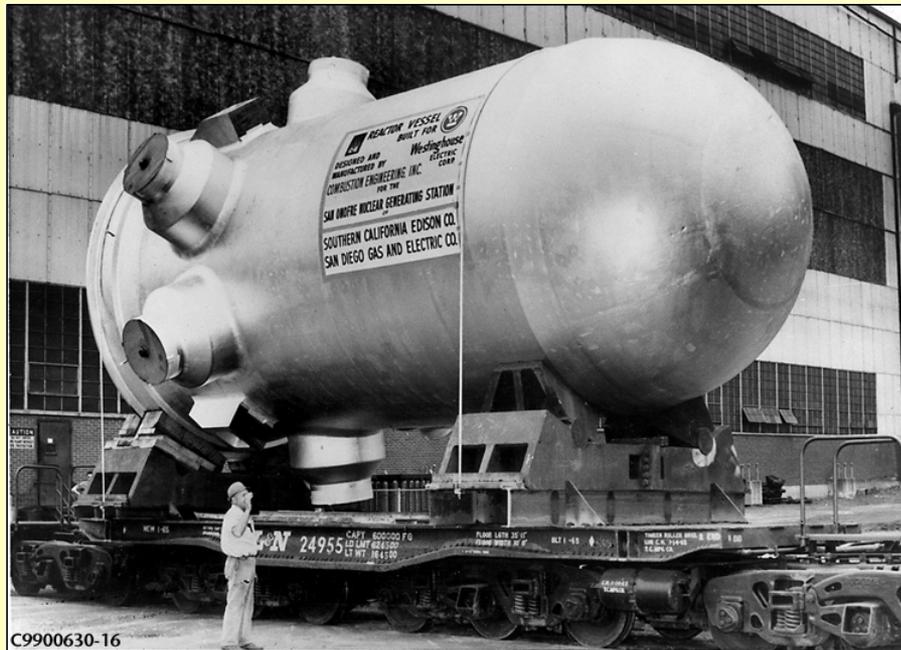
- costs for design and execution of reactor irradiations
- costs of post-irradiation examination of radioactive materials
- declining facilities for both irradiation and examination
- **combinatorial problem**: broad range of materials, phenomena, and irradiation conditions - coolants, temperature, loading conditions, dose rate, dose

Recent advances in computational hardware, computational science, ... make it more feasible than ever to aggressively attack this challenge

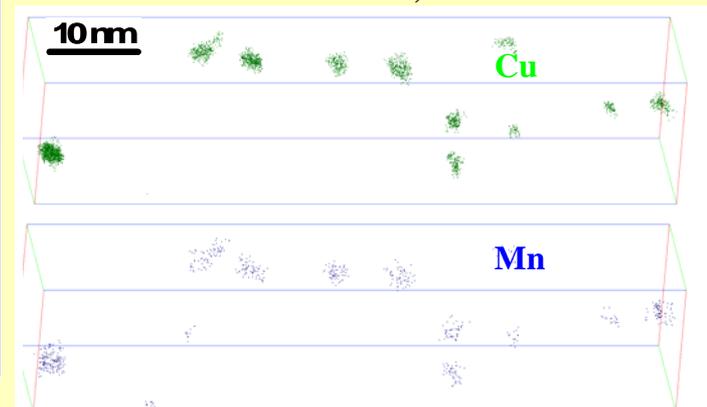
- e.g. April 2004 SC-NE Workshop on Advanced Computational Materials Science: Application to Fusion and Generation IV Fission Reactors, report at: <http://www.csm.ornl.gov/meetings/SCNEworkshop/DC-index.html>

# Understanding the Effects of Irradiation on Structural Materials Requires Multiscale Modeling (and Experiments)

- critical fracture toughness of 800 ton reactor pressure vessel can be severely degraded by radiation-induced defect structure on the size scale of 2 to 3 nm

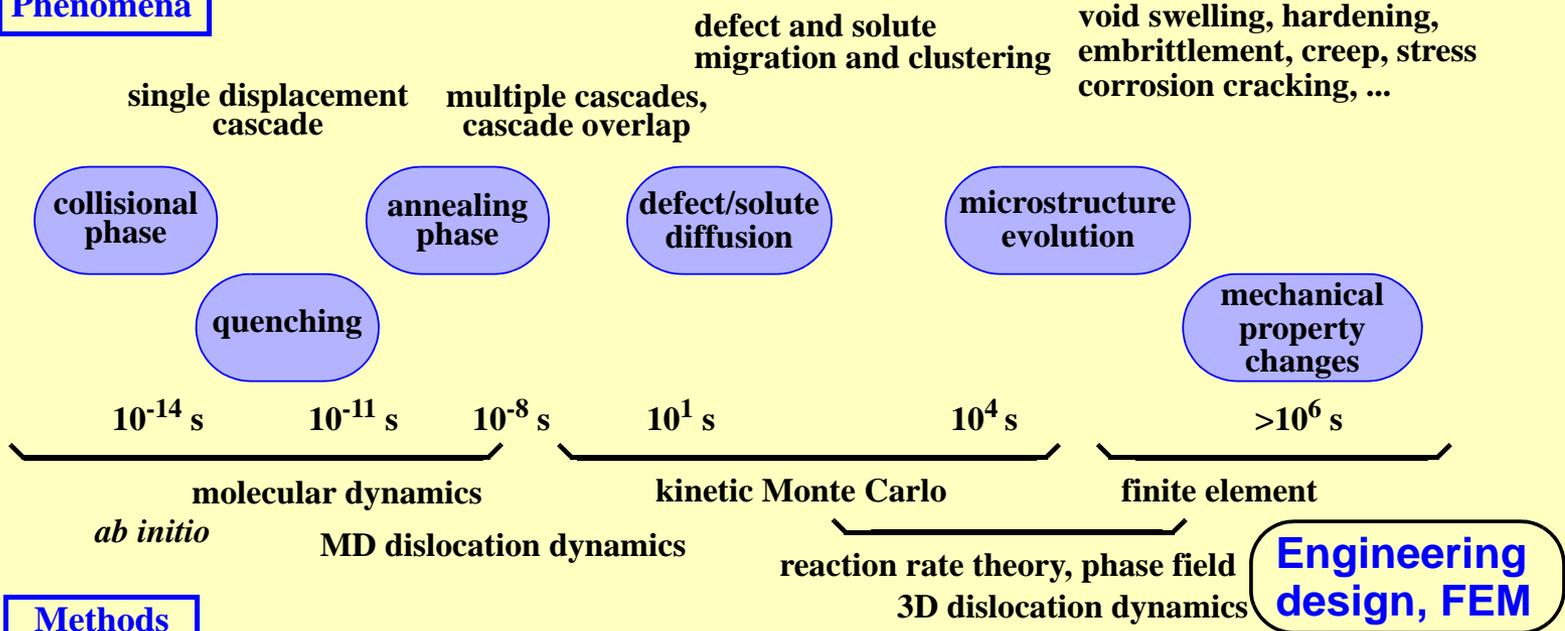


- copper and manganese-enriched clusters in neutron irradiated model RPV steel, APFIM data

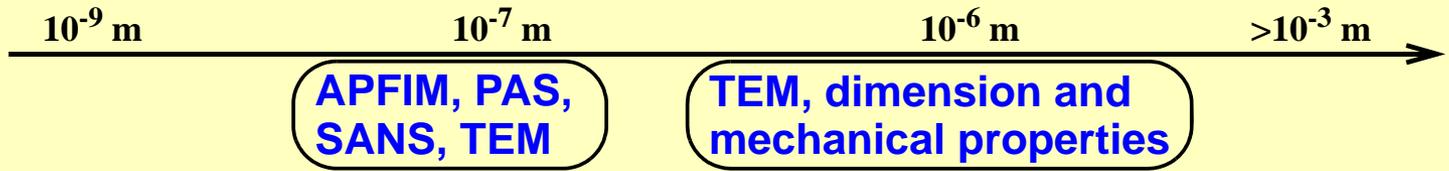


# Relevant phenomena and computational methods

## Phenomena



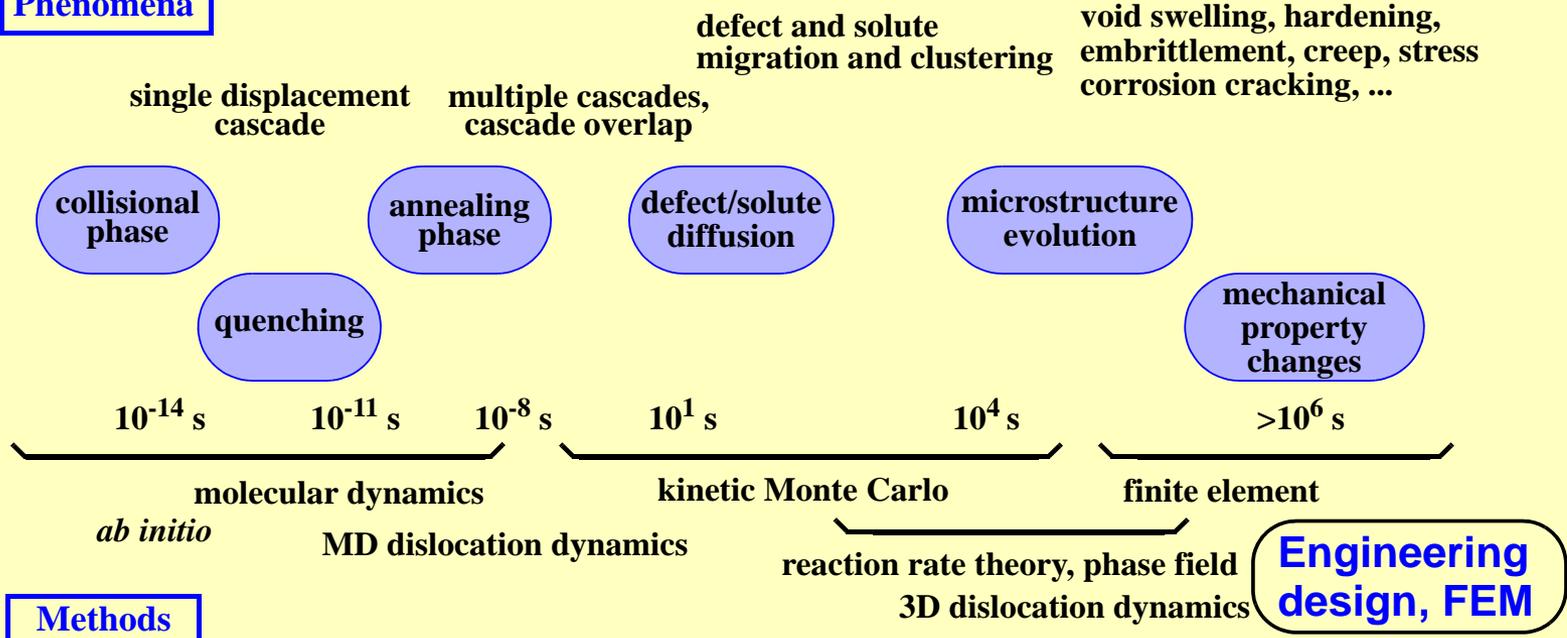
## Methods



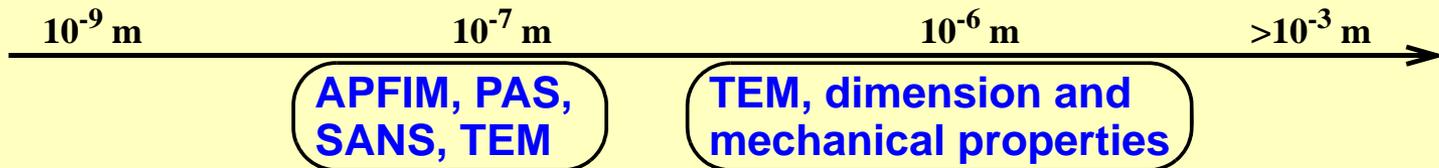
Goal of multiscale modeling is fully predictive capability, but:

# Relevant phenomena and computational methods

## Phenomena



## Methods



Goal of multiscale modeling is fully predictive capability, but: **“Prediction is very difficult, especially if it’s about the future.” ... Niels Bohr**

## Provide several brief examples from various aspects of the multiscale modeling problem:

- primary damage formation, molecular dynamics and kinetic Monte Carlo
- Helium effects in metals
  - results of early kinetic models
  - *ab initio* (VASP), accounting for He defects in iron, influence on defect properties
  - development and application of new interatomic potential
- atomistic simulation of dislocation-defect interactions, molecular dynamics
- mesoscale models of microstructural evolution
  - mean field reaction rate theory, model parameterization
  - illustrate loose (parameter passing) multiscale modeling
  - comparison of reaction rate models: mean field and KMC

# Aspects of Primary Radiation Damage Source Term

## (1) associated with fission or fusion reactions in reactors

- “fission fragments”, heavy charged particles recoiling from fission event
  - peaks around atomic masses 90 and 140
  - energy  $\sim 80\text{-}100$  MeV
  - limited range, primarily impacts fuel
- high energy neutrons (flux  $>0.1$  or  $>1.0$  MeV traditionally used as correlation parameters by nuclear industry)
  - fission spectrum, energy up to  $\sim 20$  MeV, peak at  $\sim 0.65$  MeV,  $\phi(\text{peak})/\phi(10 \text{ MeV}) \sim 350$
  - DT fusion at 14.1 MeV
  - displacement cross section minimum at  $\sim 1$  keV (elastic scattering limit) for iron
- thermal neutrons
  - typically  $E < 0.5$  eV,  $kT_{\text{room}} = 0.025\text{eV}$
  - produce low energy recoils from  $(n,\gamma)$  capture reactions; a few 100s eV in steels

## Primary Radiation Damage, con't.

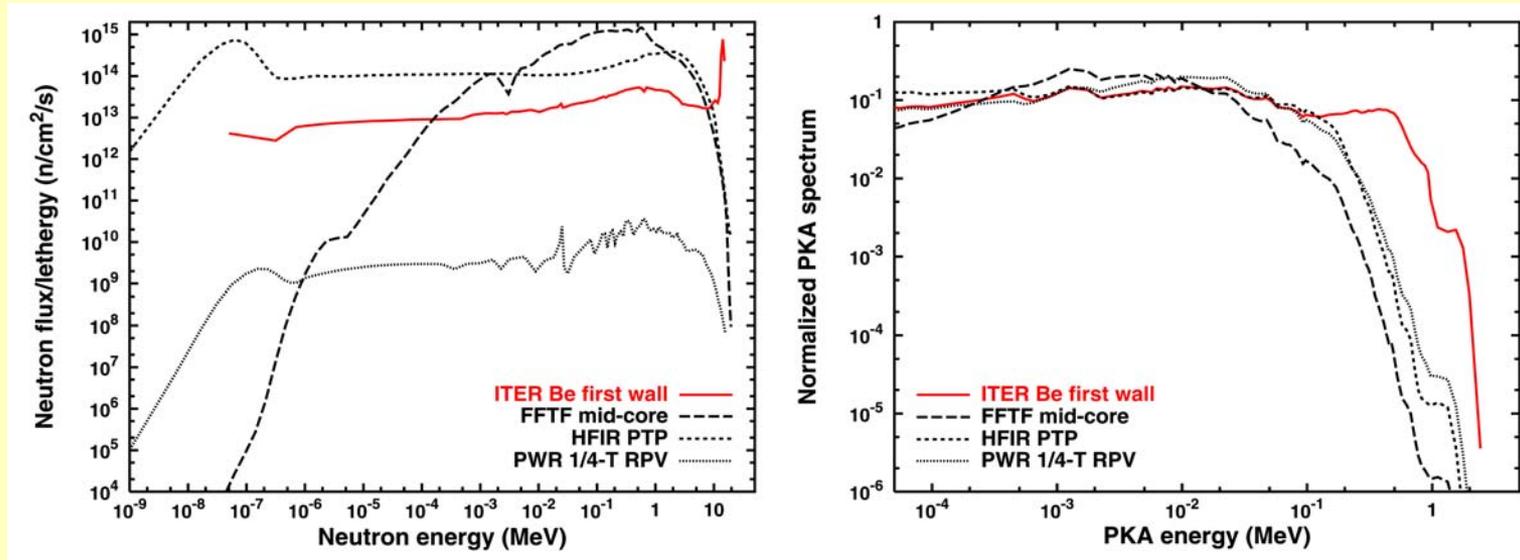
- high energy (up to a few MeV) electrons
  - primarily produced by Compton scattering of fission gamma rays, some from (n, $\gamma$ ) reactions
  - generate low energy recoils (similar to thermal neutrons) by elastic scattering
- nuclear transmutation products
  - gases: primarily hydrogen and helium from (n,p) and (n, $\alpha$ ) reactions
  - solid: (n,p), (n, $\alpha$ ), (n,2n), (n, $\gamma$ ) with subsequent  $\beta$  decay
  - both thermal and high energy neutron reactions contribute
  - appm to atom-% levels, generally not too significant, but e.g. silicon production in aluminum where  $\phi_{th}=2.5 \times 10^{26}$  n/m<sup>2</sup> (~6 months in HFIR) converts 1% of Al to Si

## Primary Radiation Damage, con't.

### (2) accelerator based sources

- light and heavy ions
  - a few 100 keV to 5 MeV typical (up to ~40 MeV in cyclotrons)
- electrons, typically 1-5 MeV
  - used for illumination and damage production in HVEM *in situ* studies
- both ions and electrons have been heavily used in radiation damage “simulation” studies, combined ion-beam/TEM facilities
- modern spallation sources with proton energies ~1 GeV
  - substantial damage from primary proton beam
  - produce neutrons with energies up to nearly the beam energy
  - the periodic table of transmutation products, primary light elements with high levels of H and He
  - radiation effects research needed to predict performance of target materials and may be useful for some fusion materials investigation

# Comparison of representative neutron and corresponding PKA energy spectra



- differences in neutron flux level lead to different atomic displacement rates
- neutron energy spectrum differences lead to different PKA energy spectra,
  - different coolants, water for HFIR and PWR vs. sodium for FFTF alter neutron energy spectrum, primarily influence lower energy
  - high energy influenced by neutron source, c.f. all fission with ITER fusion

# MD simulation of primary damage formation

Classical molecular dynamics, typical implementations:

- constant pressure, periodic boundary condition
- boundary atoms not damped, results in some heating
- no electronic losses or electron-phonon coupling, energy of cascade simulation, hence for:  $T_{\text{dam}} = \text{kinetic energy lost in elastic collisions}$ ,

$$E_{\text{MD}} \sim T_{\text{dam}} (\text{NRT}) < E_{\text{PKA}}$$

Range of interatomic potentials have been employed, from simple pair potentials to embedded atom or Finnis-Sinclair, limited work with higher order potentials

- results presented here for modified version of Finnis-Sinclair iron
- can be compared with simple standard model for number of defects produced:

$$v_{\text{NRT}} = \frac{0.8 \cdot T_{\text{dam}}}{2 \cdot E_d}$$

## Partial Fe Cascade Database at 100K

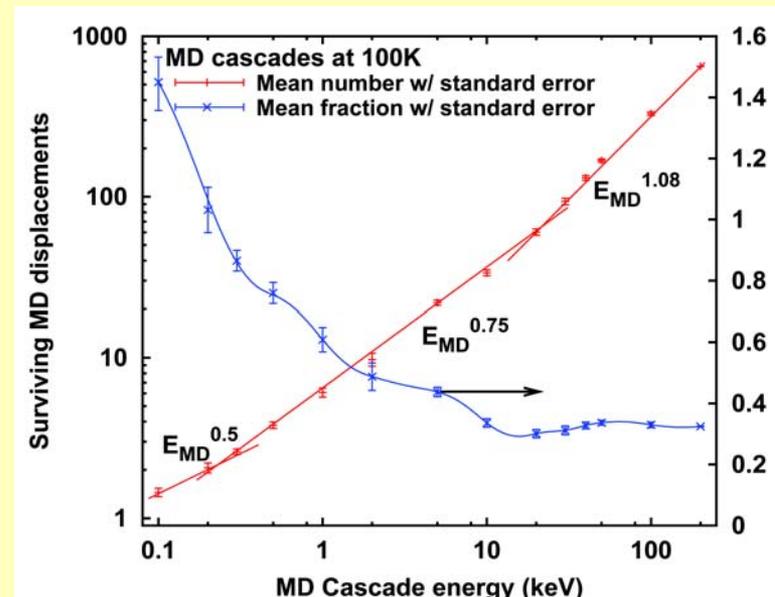
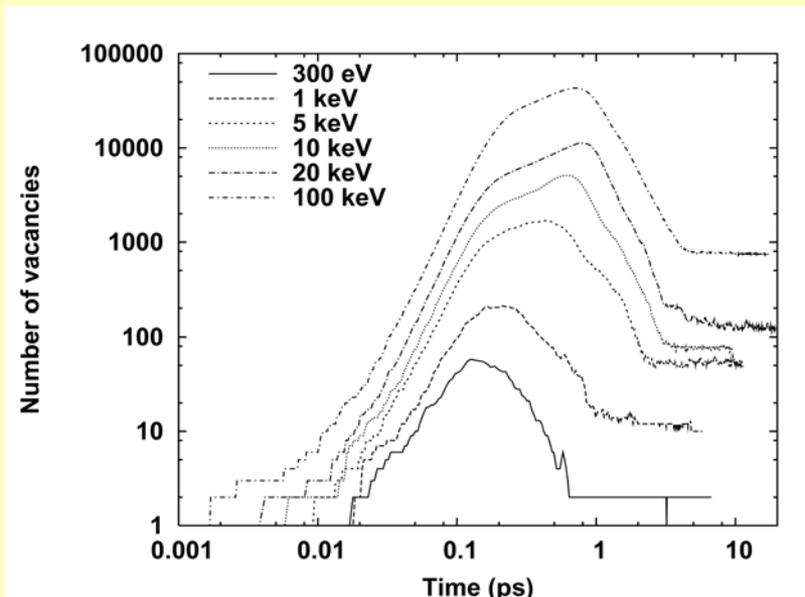
MD Cascade Energy (keV)	Corresponding neutron energy (MeV)	Typical simulation cell size (atoms)
0.1	0.003351	3,456
0.2	0.006818	6,750
0.5	0.01749	16k/54k
1.0	0.03578	54k
2.0	0.07342	54k
5.0	0.1911	128k
10.	0.3968	125k/250k
20.	0.8321	250k
50.	2.277	2.249M
100.	5.085	5.030M
200.	12.31	9.826M

# Primary damage event evolves on ps time scale

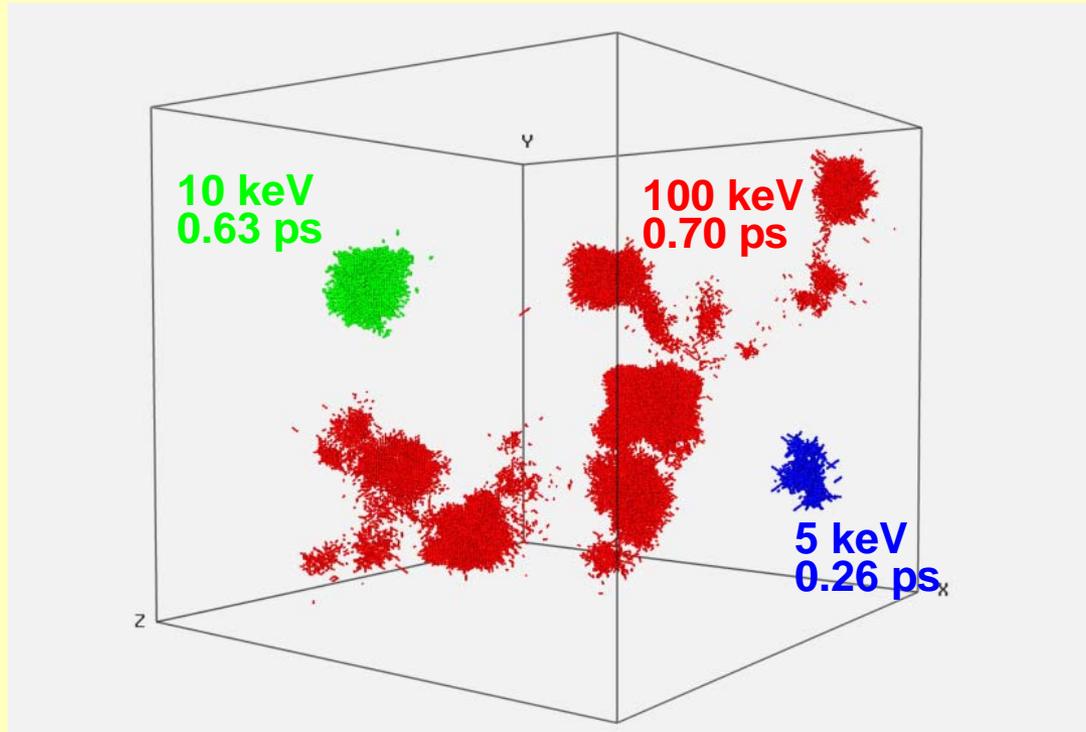
In contrast to linear damage energy dependence of NRT model, three well defined energy regimes appear

- at lowest energies true “cascade-like” behavior does not occur
- above ~10 keV, subcascade formation dominates
- nearly linear energy-dependence is observed at higher energies, consistent with simple reasoning of K-P or NRT models

50 keV  
example

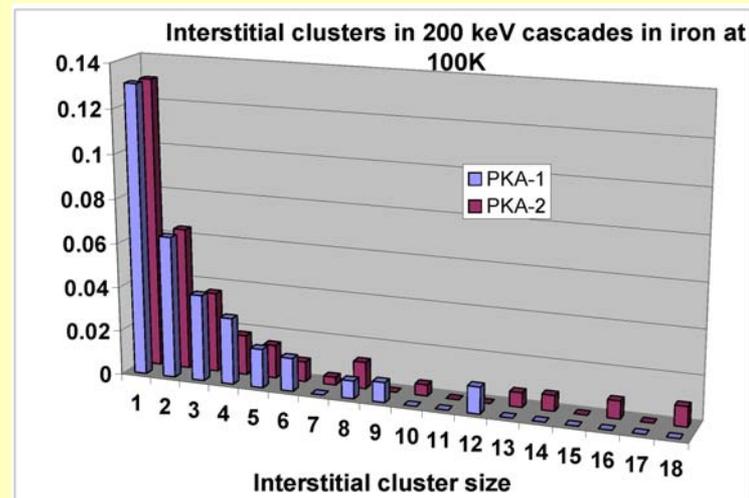
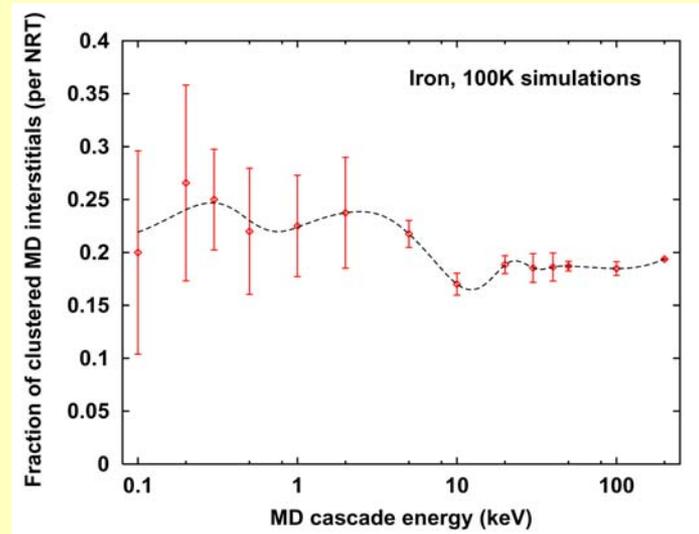
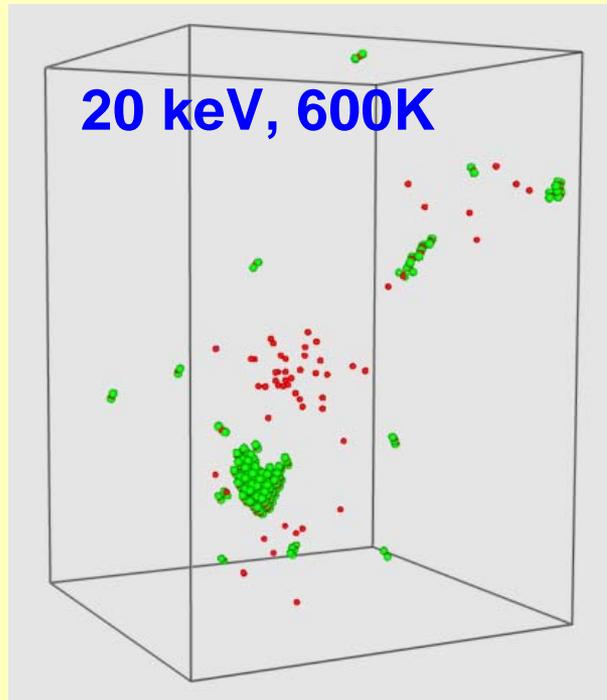


## Illustration of subcascade structure at peak damage condition for 5, 10, and 100 keV cascades at 100K



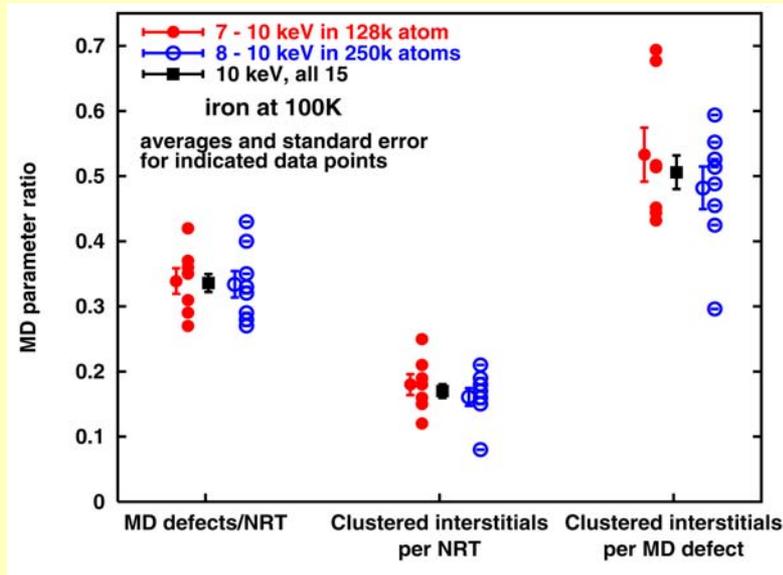
- high energy cascades look like multiple lower energy events, leads to asymptotic behavior with energy
- low energy events between subcascades have higher efficiency

Primary damage production involves both isolated Frenkel pair and clusters of point defects - significant impact on nucleation

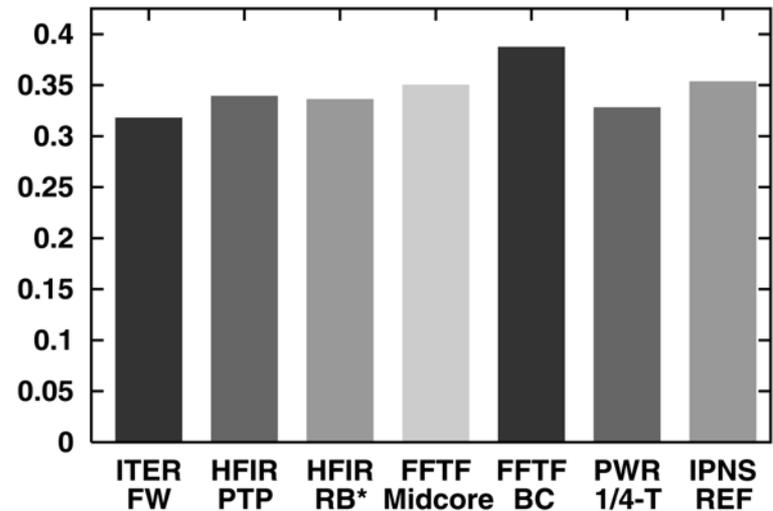


## Cascade Statistics and Averaging

- statistical character requires “enough” simulations at any one condition
- account for neutron energy spectrum by averaging over PKA spectrum



### Spectrum-averaged: total defect survival (per NRT)



# Status of Primary Radiation Damage Simulation

- extensive MD cascade studies have been carried out, largest database for Ferris-Sinclairium “iron”
- analysis of the simulations has lead to a good mean description of the dependence on temperature and cascade energy up to fusion-relevant energies
- some anticipated and new phenomena have been revealed and explained: subcascade formation, glissile interstitial cluster formation, 3D and planar channeling effect

## Needs

- many more simulations needed to determine influence of ‘rare’ events
- longer times to be explored in cascade aging studies using KMC
- potential need for more realistic potentials for transition metals such as iron to account for partially-filled d-bands (directional effects) and magnetism
- realistic potentials for multicomponent alloys: ~5 major components in most steels plus critical effects of minor solutes and impurities

## Influence of Helium on Microstructural Evolution

- Helium and hydrogen can be produced in significant quantities by  $(n,\alpha)$  and  $(n,p)$  reactions in most materials
- Helium and residual gases were implicated in the formation of cavities in irradiated steels in the late 1960s and early 1970s
- He generation rate a strong function of neutron energy spectrum and material composition, threshold reactions and thermal reactions
- A particular concern for nickel-containing alloys exposed to thermal neutrons and all materials in in DT fusion environment
  - stainless steel in fast reactor,  $\sim 0.5$  appm/dpa
  - stainless steel in thermal reactor, non-linear  $\sim 1$  to 70 appm/dpa
  - stainless steel in DT fusion reactor,  $\sim 10$  appm/dpa

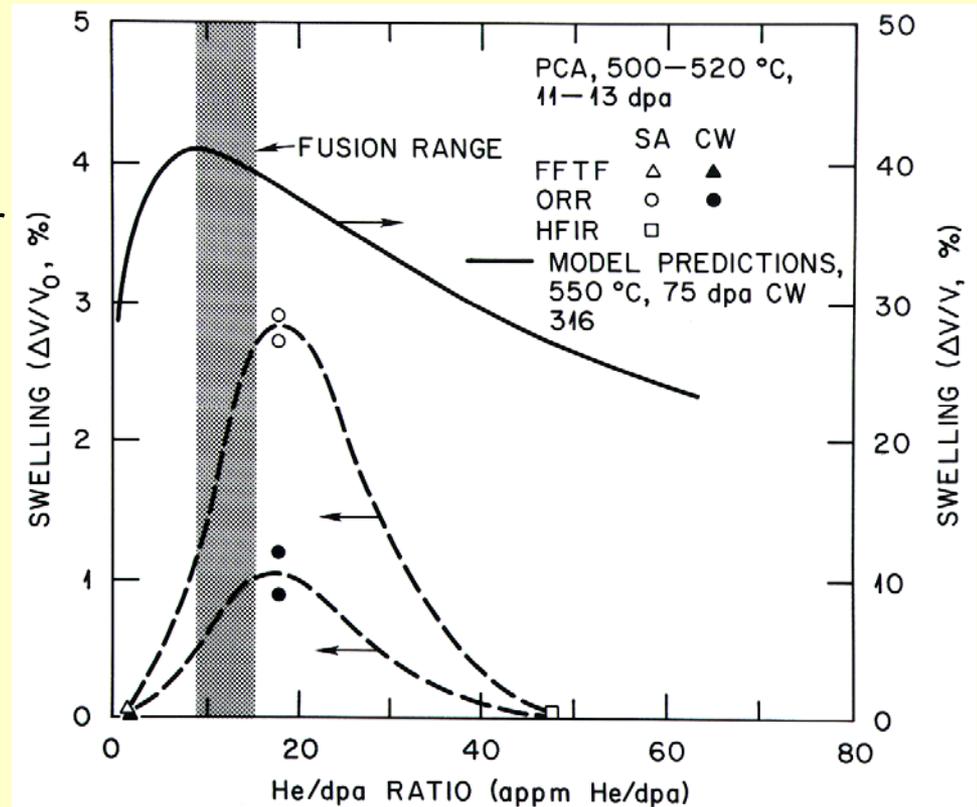
# Early mechanistic models used to anticipate role of transmutant helium on swelling

- Helium effects investigated
  - experiments with isotopic tailoring of Ni in austenitic steel used to obtain intermediate and high helium in water moderated reactors (ORR and HFIR)
  - experiments confirmed model predictions of possible non-monotonic behavior

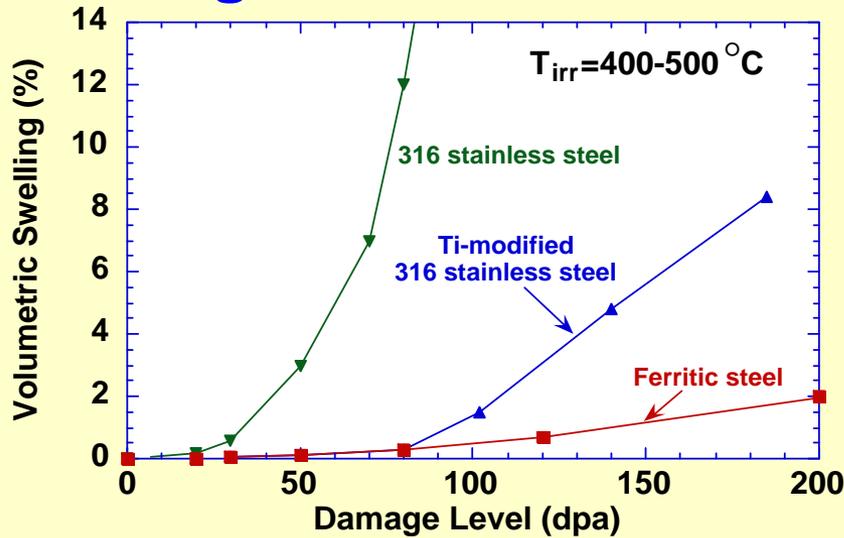
Some success, but confidence limited by nature of models and parameters, e.g.

- basic material and defect parameters, point defects and helium
- nucleation model
- ...

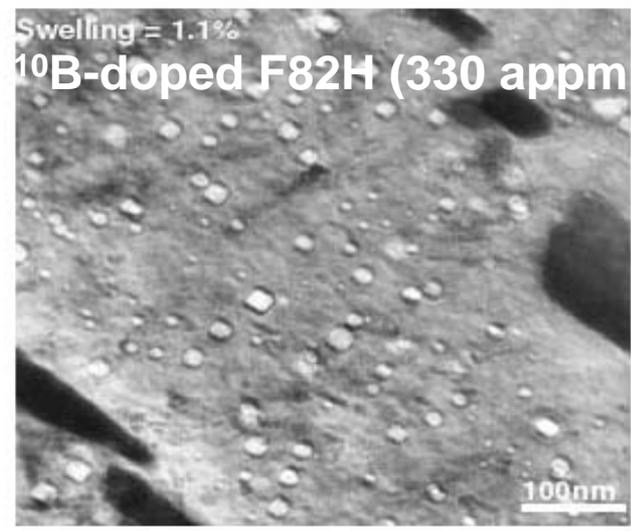
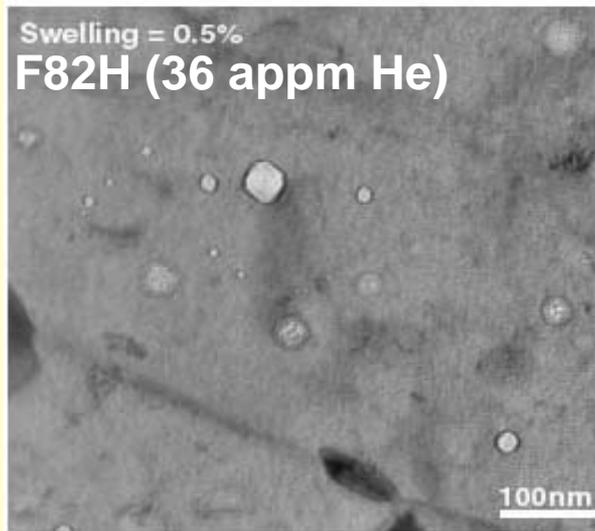
Subsequent progress addressed primary damage formation, fundamental behavior of He, cavity nucleation, He transport



# Swelling can eliminate materials from consideration



**Current fusion candidate alloys not immune to He effects:  
HFIR irradiation at 400°C to 51 dpa, *E. Wakai, et al. JNM (2000)***

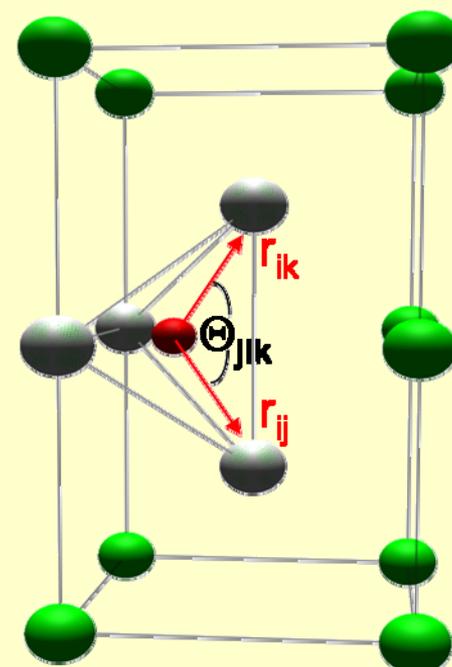


# New three-body He-Fe potential

- Fitted to VASP calculations of both relaxed and unrelaxed defects
  - Both forces and energies fitted
  - Large defects such as  $\text{He}_3\text{V}$  calculated with up to 128 atoms
- Must predict tetrahedral site as most stable site for He
  - Difficult with pair or EAM functional forms, so 3-body form used
- Potential consists of a repulsive pair term  $\phi$  and a 3-body term  $Y$

$$E = E_{\text{Fe-Fe}} + E_{\text{He-He}} + \sum_{\substack{i \in \text{He} \\ j \in \text{Fe}}} \phi(r_{ij}) + \sum_{\substack{i \in \text{He} \\ j, k \in \text{Fe}}} Y(r_{ij}, r_{ik}, \Theta_{jlk})$$

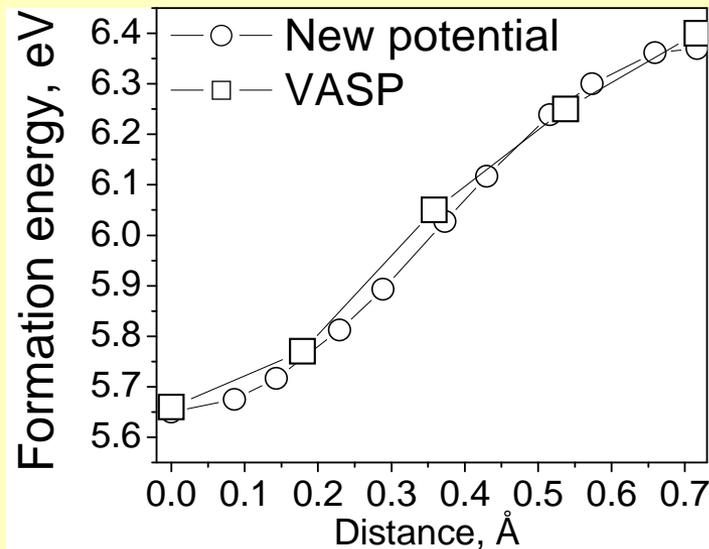
- Considerable effort invested to properly understand and implement 3-body potential
- Fe-Fe matrix can be:
  - Finnis – Sinclair pair potential (1984)
  - Ackland – Bacon EAM potential (1997)
  - Ackland – Mendeleev EAM potential (2004)
- He - He is Aziz pair potential (1995)



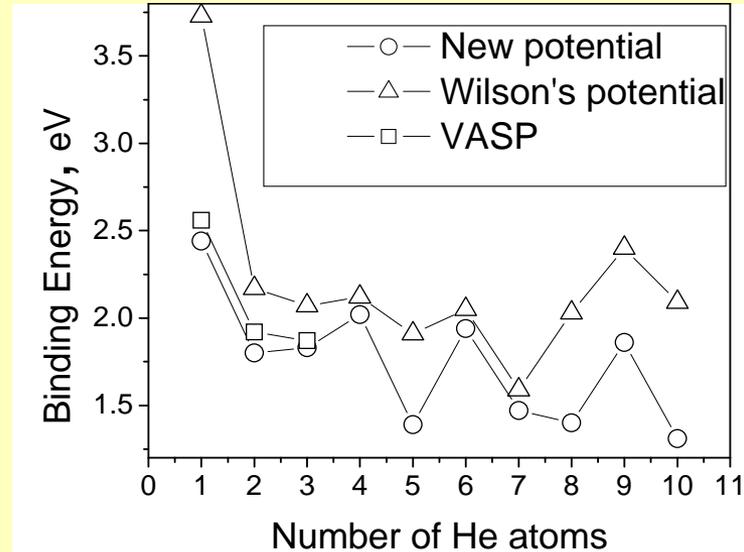
## Fundamental behavior of helium

- tetrahedral site stabilized by unexpected hybridization of Fe-He electron orbitals and small affect on Fe magnetic moment
- comparison of new potential with VASP results
  - use of many-body He-Fe potential based on Finnis-Sinclair Fe shown in (a)
  - He-vacancy cluster binding energies found to be substantially different than earlier work (b)

(a) He interstitial formation energy along (100) path from tetrahedral to octahedral

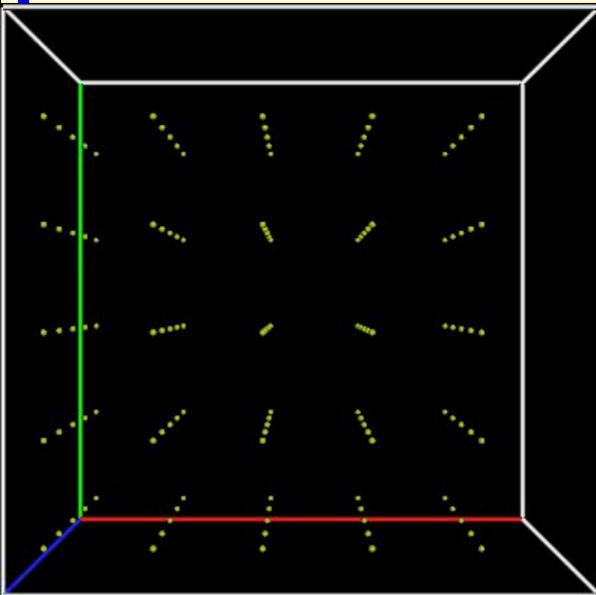


(b) binding energy of helium to single vacancy



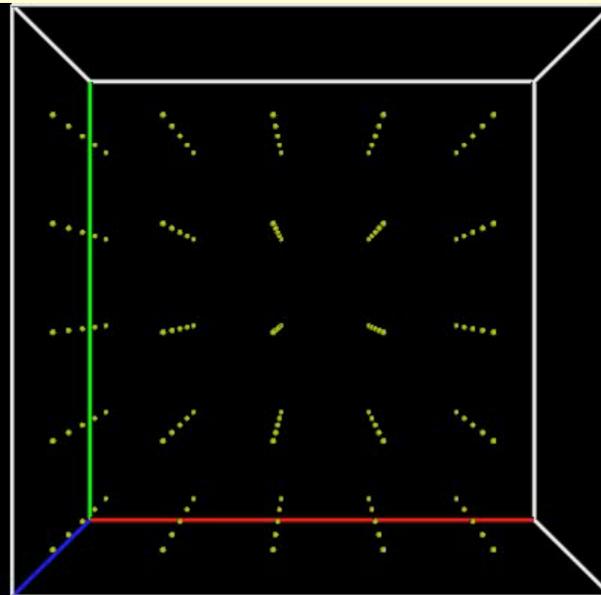
# Extensive investigation of He and He-vacancy clusters

- He diffusion and clustering properties
- Behavior of different He-Fe potentials
  - Wilson potential creates FP (SIA emission) more quickly than ORNL potential, higher He binding energy.
  - Juslin-Nordlund potential does not form He clusters or create FP above  $\sim 400^{\circ}\text{C}$



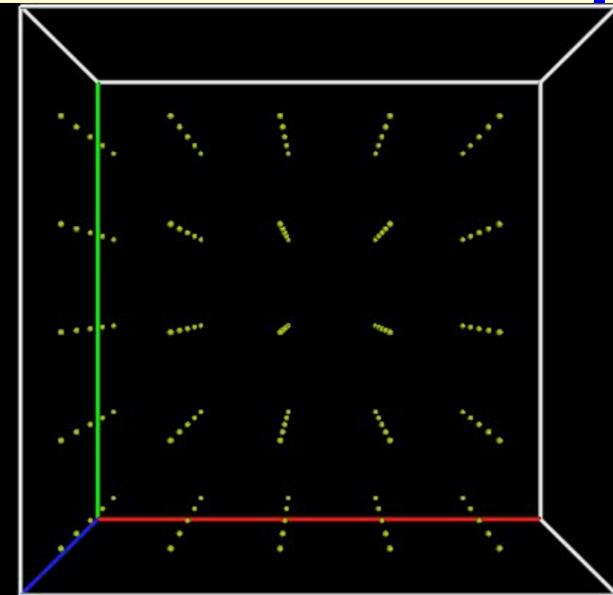
Wilson potential

0ps 600K 2089appm He



ORNL potential

Ackland 97 Fe potential



J-N potential

# Applications for Fe-He potential

- development of theory-based EOS for He in Fe, for application in mesoscale and continuum models
- MD-based investigation of He-vacancy defects
- use MD simulations to provide parameters for cluster dynamics models to investigate evolution of He-vacancy clusters leading to bubble nucleation

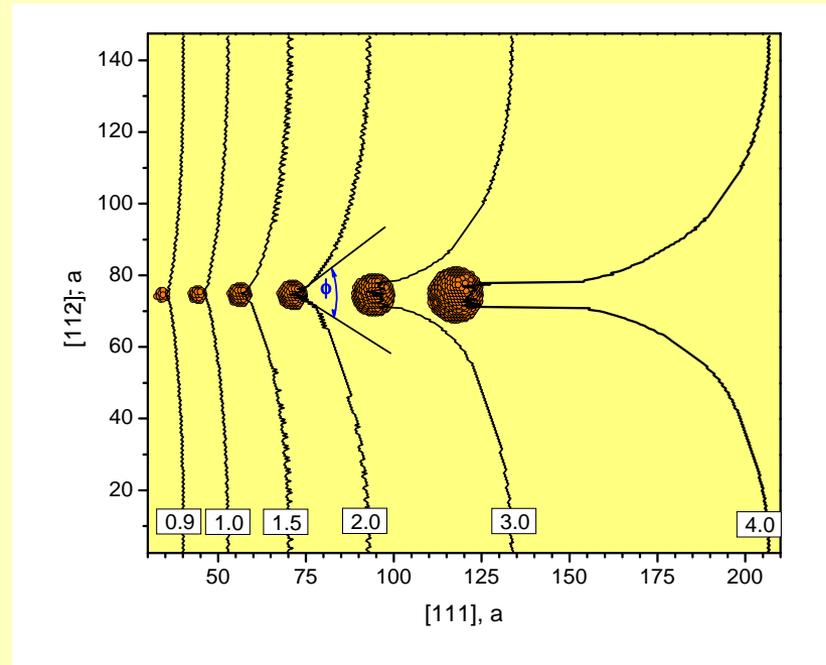
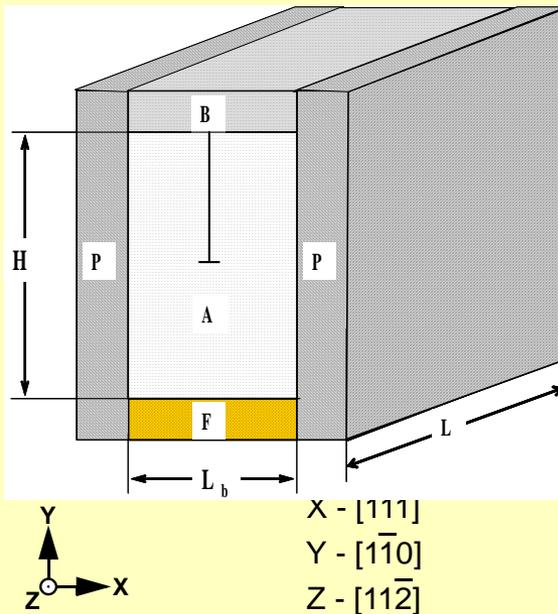
## Needs

- scaling up of *ab initio* calculations to examine larger defects
- verification of DFT results using e.g. quantum Monte Carlo
- as above, additional validation of simple embedded atom type potentials
- similar work needed for other interesting metals and alloys
- effects of hydrogen need similar investigation

# Applications of MD-based dislocation dynamics

Investigate atomistic details of dislocation-defect interactions, relevant to hardening mechanisms: dislocation loops, voids, SFTs, precipitates

- obtain critical resolved shear stress as a function of dislocation density, defect density and size; influence of temperature and strain rate
- identify mechanisms important in clearing defects and forming defect-free channels, important in flow localization



# Applications

## (1) Atomistic nature of dislocation-defect interactions

- radiation-induced increase in yield strength based on simple hardening theory for dispersed barriers

$$\Delta\sigma = T\Delta\tau = T(\alpha Gb) \cdot (\sqrt{Nd})$$

where T is the Taylor factor (3.06), N and d are the radiation-induced defect density and mean diameter, and  $\alpha$  is the so-called barrier strength

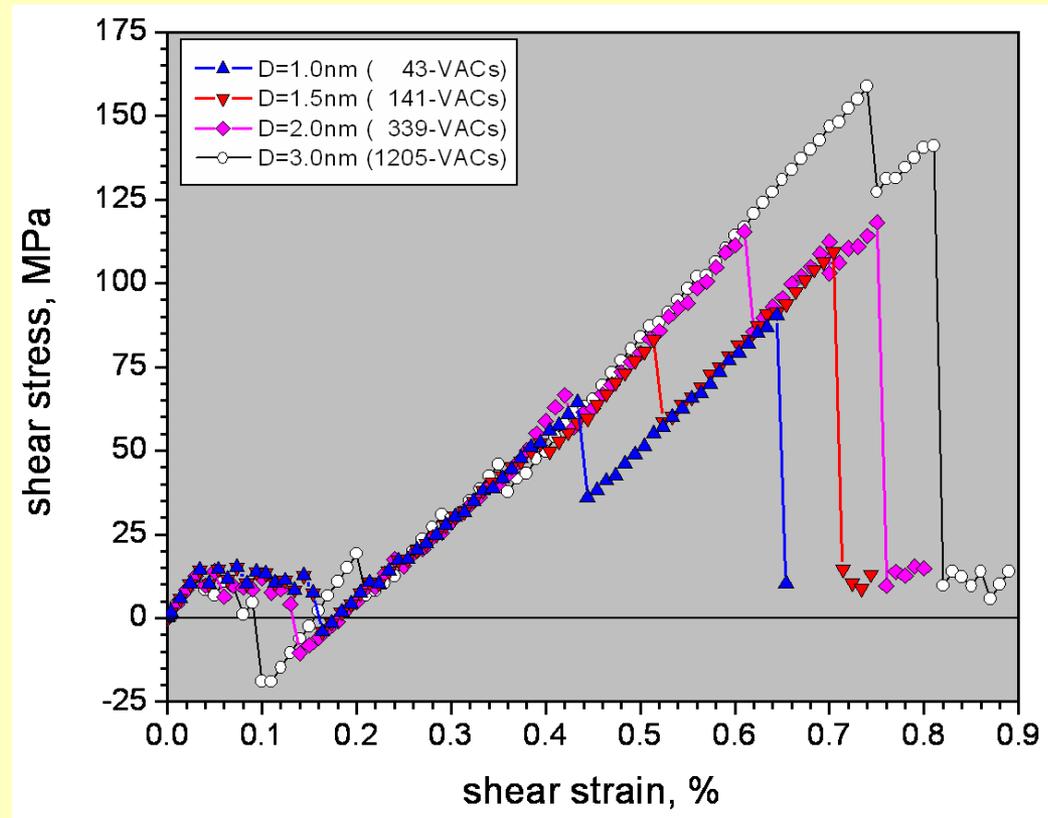
- values of the barrier strength can be estimated from continuum elasticity calculations and from comparisons between microstructural observations (TEM, APFIM, SANS) and mechanical property changes
- typical values are 0.1 to 1.0, considerable uncertainty associated with superposition rules for multiple defect types, error in microstructural measurements, allowance for invisible defects, etc.

Fe Void Example-1

Cu SFT Example-2

## Elasticity-based dislocation models can not account for atomistic details of dislocation-defect interactions

- void shearing, precipitate shear and phase transformation (not shown)
- effects of dislocation dissociation into partials



## 2) Formation of nearly defect-free channels

- Investigation of defect absorption mechanisms and relevance to comparison between bulk and thin film experiments
- Nb irradiated to neutron fluence ( $E > 1.0$  MeV) of  $4.4 \times 10^{22}$  n/m<sup>2</sup> at  $\sim 50^\circ\text{C}$  and strained to 6% at room temperature

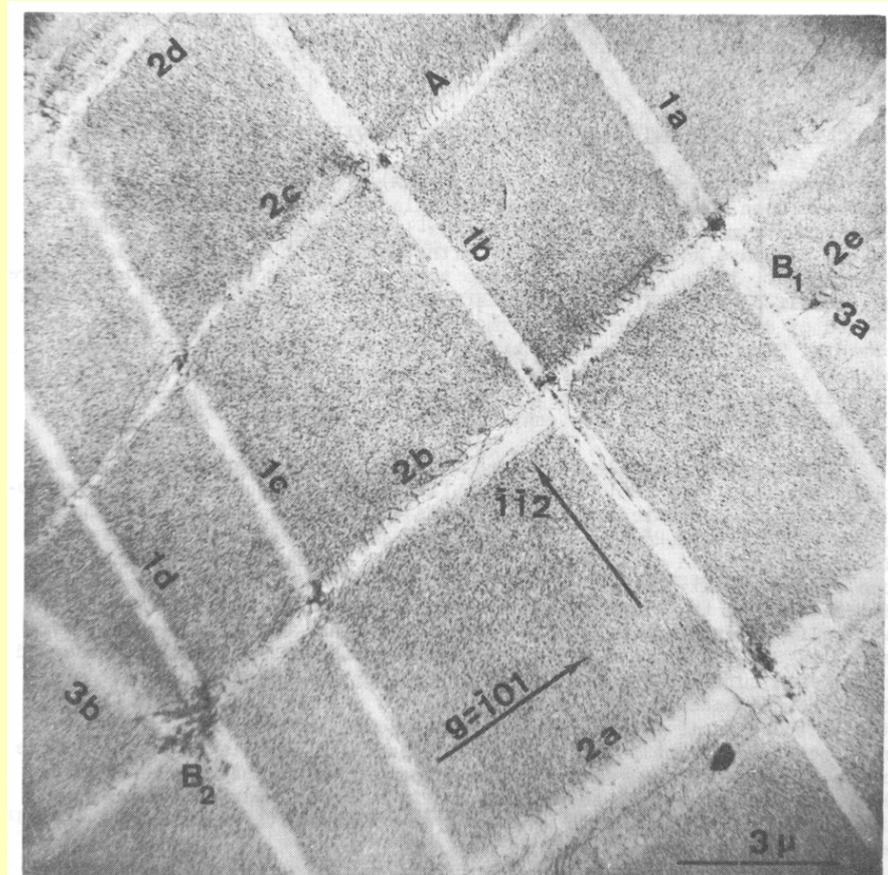


FIG. 5. Rectangular pattern of dislocation channels in niobium irradiated to  $4.4 \times 10^{18}$  n/cm<sup>2</sup> ( $E > 1$  MeV) at  $50^\circ\text{C}$  and strained to 6.6% at room temperature.

Cu SFT Example-3

# Status of MD Dislocation Dynamics

- relevance of atomistic simulations clearly demonstrated, e.g. dislocation-point defect reactions leading to climb, jog formation, etc.; defect destruction, defect creation (single and clusters), effect of free surfaces
- extensive investigations have included edge and screw dislocations in Fe and Cu with obstacles including precipitates (Cu in Fe), SFTs, dislocation loops, and voids, work in progress for multiple defect types and He-filled bubbles
- variables include strain rate (dislocation velocity), temperature, effective dislocation and obstacle densities
- importance of matching experimental conditions to simulations

## Needs

- more and faster cycles to enable more experimentally relevant strain rates, and defect sizes and densities
- hybrid models linking MD up to continuum and down to *ab initio*, Green's function boundary conditions employed in some cases but more work needed

# Application of mesoscale models in radiation effects

- mesoscale models are relevant to many phenomena in materials science and radiation effects
  - grain growth
  - dislocation evolution, by thermo-mechanical or radiation-induced processes
  - void swelling
  - precipitation of additional phases, and solute segregation
  - stress corrosion cracking, and irradiation-assisted SCC
- size scale permits direct comparison with experiments such as TEM and mechanical property measurements
- dependent on fundamental atomistic processes, and controls macroscopic observables such as strength, ductility, creep, ...
- a primary application is the investigation of point defect and solute kinetics and microstructural evolution, so-called mean field models based on reaction rate theory, phase field models and some types of Monte Carlo simulations

## Example from mean field reaction rate theory modeling

starting point is continuity equations describing point defect (vacancy,  $C_v$  and interstitial,  $C_i$ ) populations (analogous equations for solutes):

$$\nabla \cdot \left( D_v \nabla C_v + \frac{D_v C_v}{kT} \nabla U_v \right) + G_v - \alpha C_i C_v - D_v C_v S_v^T = \frac{\partial C_v}{\partial t}$$

$$\nabla \cdot \left( D_i \nabla C_i + \frac{D_i C_i}{kT} \nabla U_i \right) + G_i - \alpha C_i C_v - D_i C_i S_i^T = \frac{\partial C_i}{\partial t}$$

where the  $\nabla$  denote spatial derivatives.

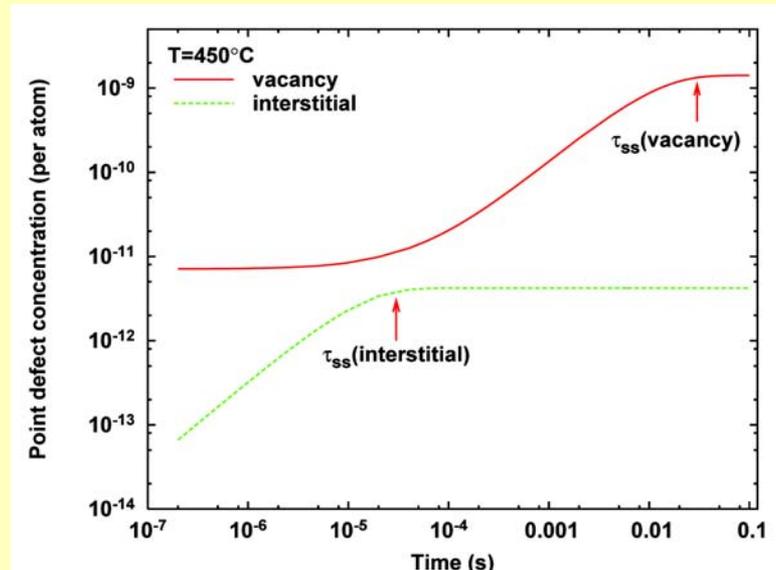
- first term on the LHS describes point defect drift to discrete sinks, the  $U_{i,v}$  are interaction energies between the point defects and discrete sinks
- $G_{i,v} = \eta G_{dpa} + G_{i,v}^{em}$  is the total point defect generation rate, including thermal emission from sinks, and the  $D_{i,v}$  are the point defect diffusivities
- $S_{i,v}^T$  are the total sink strengths for continuum sinks (e.g. cavities, dislocation, grain boundaries, etc.), recombination rate coefficient is given in terms of an effective recombination radius,  $r_r$ :  $\alpha = 4\pi r_r (D_v + D_i)$

- typical assumptions/simplifications
  - the material is treated as a spatially-homogeneous effective medium with embedded effective sinks and sources for point defects
  - spatially-averaged point defect generation rates are also generally employed
  - these assumptions have been relaxed in particular cases, e.g. to investigate cascade-induced fluctuations in point defect concentrations
  - the models are formulated as a series of differential equations describing the production and fate of point defects and the corresponding evolution of the microstructure
- With these approximations, and assuming that the irradiation produces only monomers, the time-dependent or steady state point defect concentrations can be obtained as a solution to the following equations:

$$\frac{dC_{i,v}}{dt} = \langle \eta G_{dpa} + G_{i,v}^{em} \rangle - \alpha C_i C_v - D_{i,v} C_{i,v} S_{i,v}^T$$

## Time dependent solution of simple point defect equations

- much different time constants for vacancies and interstitials due to much slower vacancy migration



- a minor multiscale problem within a mesoscale method, requires good ODE solver for stiff systems

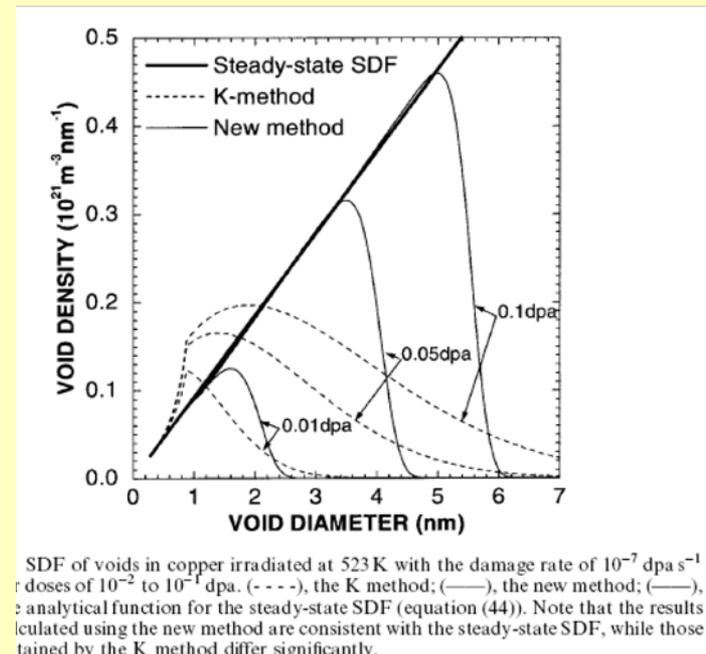
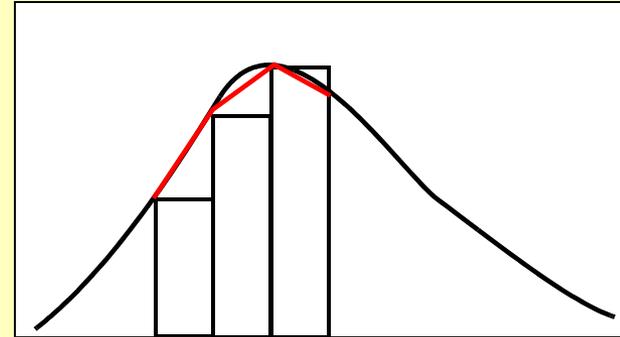
- analogous equations can be written to describe an evolving point defect cluster population, for helium generation and distribution, and for the other microstructural components, e.g.:
  - if only the monomers are assumed to be mobile, an equation describing the di-interstitial population,  $C_{2i}$ , can be written:

$$\frac{dC_{2i}}{dt} = \beta_i^j C_i^2 + \beta_v^{3i} C_v C_{3i} - C_{2i} (\beta_i^{2i} C_i + \beta_v^{2i} C_v)$$

- this equation can be generalized to formulate a master equation for interstitial (and vacancy) clusters of arbitrary size. However, this also generates an arbitrarily large number of equations - smallest visible defect clusters ~100 point defects
- various grouping schemes have been devised to minimize number of equations, introduce potential source of error
- modern computers reduce driving force for limiting number of equations, improved methods still being developed

## Modern Method for Discretizing Master Curve

- traditional methods amount to rectangular approximation in integration, fail to simultaneously conserve both the total number of (e.g. void density) clusters and the number of point defects in clusters (e.g. swelling)
- new grouping method amounts to a trapezoid approximation, does conserve both quantities - see Ovcharenko, Golubov, et al., *Comp. Phys Comm.* 152 (2003) 208.
- Comparison with analytical results: homogeneous nucleation
  - traditional method does not match analytical solution at high dose
  - new method (Golubov, et al., *Phil. Mag. A* 81 (2001) 643)



## ... back to rate equations

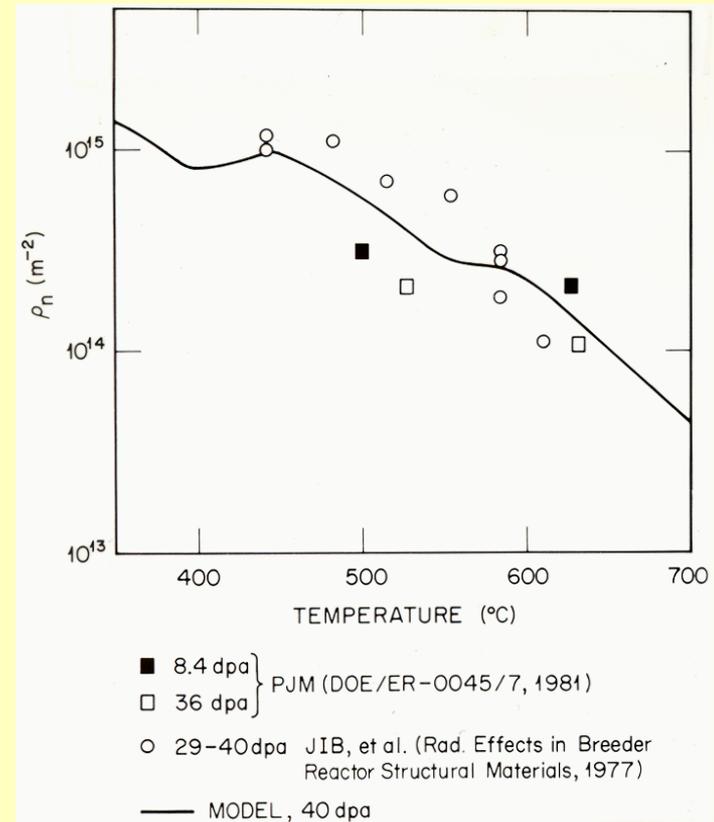
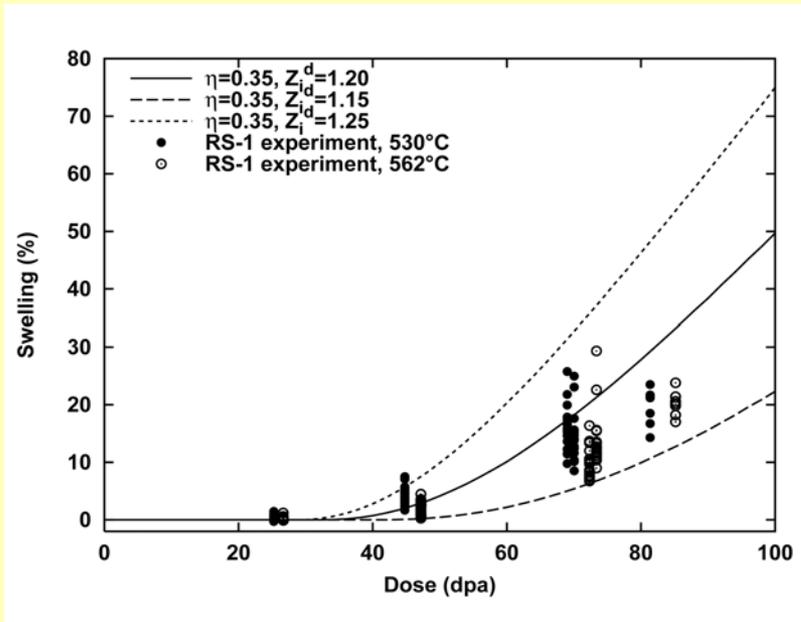
- analogous rate equations can be written to describe the evolution of extended defects, void growth rate and void sink strength:

$$S_{i,v}^v = Z_{i,v}^v 4\pi r N_v (1 + r_v (S_{i,v}^T)^{0.5})$$

$$\frac{dr_v}{dt} = \frac{1}{r_v} \left( Z_v^v D_v (C_v - C_v^v) - Z_i^v D_i C_i \right)$$

- analogous expressions available for grain boundaries, dislocations, dislocation loops, etc.
- greater or lesser detail can be built in as needed to simulate given phenomena e.g. defect nucleation vs. growth regimes, dislocation evolution, effects of solute segregation, etc.

- solutions obtained simultaneous integration of equations included in a given model and, when well calibrated with experimental data, such models have some predictive capability

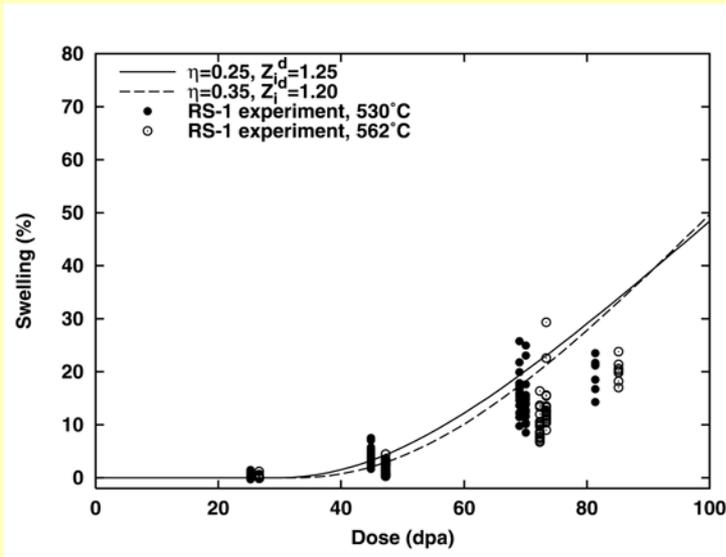


However, the success of these models in fitting data can be deceiving (the devil is in the details ...)

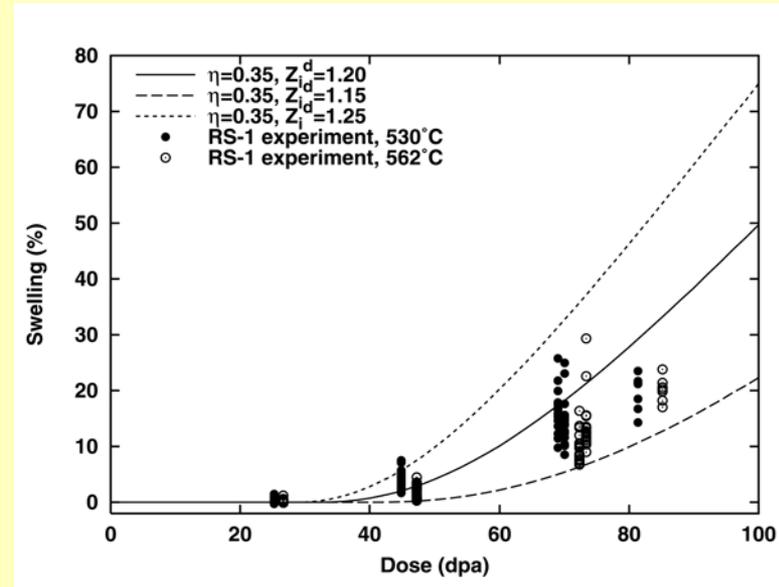
- data fitting with incomplete models leads to use of “effective” parameter values, use of parameter-rich models limits confidence in model extrapolation
- for example, if point defect absorption dominated by dislocations with sink strength  $S_d$ , swelling rate is proportional to product of  $\eta$  and net dislocation bias ( $Z_i^d - 1$ ):

$$\frac{dV}{dt} = \frac{\eta G dpa}{S_v^d} (Z_i^d - 1)$$

- In this case, data fitting can not be used to obtain unique set of model parameters. MD cascade simulations provide independent estimate of  $\eta$  and thereby permit better estimate of  $Z_i^d$ .



- arbitrary  $Z_i^d$  and  $\eta$



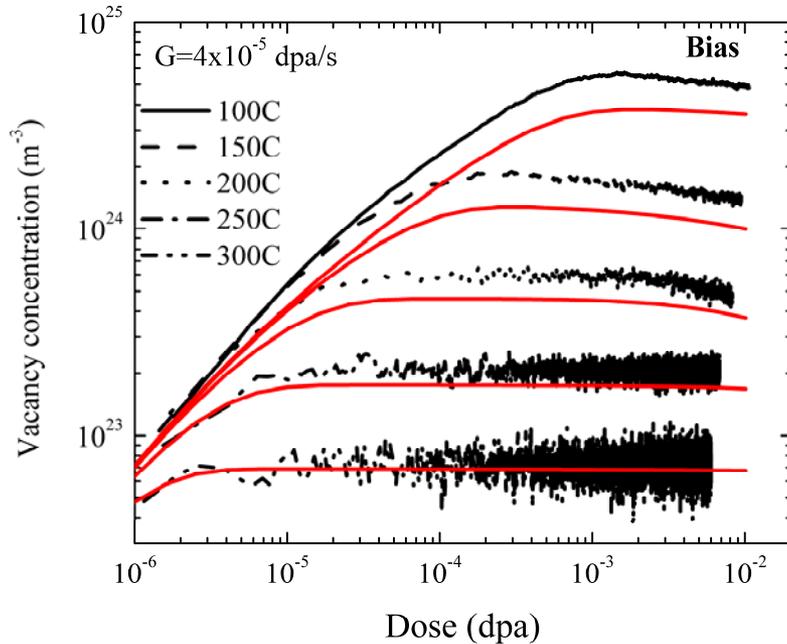
-  $\eta$  fixed by MD results,  $Z_i^d$  fit to data

- more complex models may be ‘stiffer’ with respect to arbitrary parameter choices, but more complex models introduce additional parameters

# Comparison of Alternate Mesoscale Models

- Provides additional opportunity to validate models, e.g. at mesoscale where mean field reaction rate theory (RT), Monte Carlo (MC), or phase field models are often used
- RT has a long history in radiation effects modeling; MC finding increasing use due to recent advances in computational power have expanded the domain MC models. Focus here on object kinetic Monte Carlo (OkMC)
- RT and OkMC models are similar kinetic models
  - can be used simulate the same phenomena
  - many details are handled differently in the two approaches
- A direct comparison of the RT and OkMC models
  - point defect cluster dynamics modeling, relevant to nucleation and evolution of radiation-induced defect structures
  - illustrate relative strengths and weaknesses of the two approaches

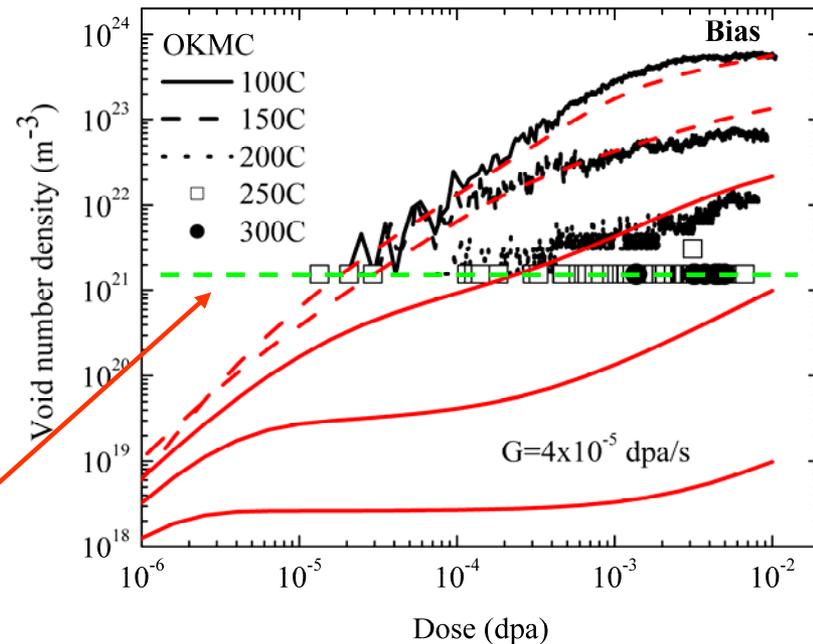
# Effect of Irradiation Temperature: $G_{\text{dpa}} = 4 \times 10^{-5} \text{ dpa/s}$



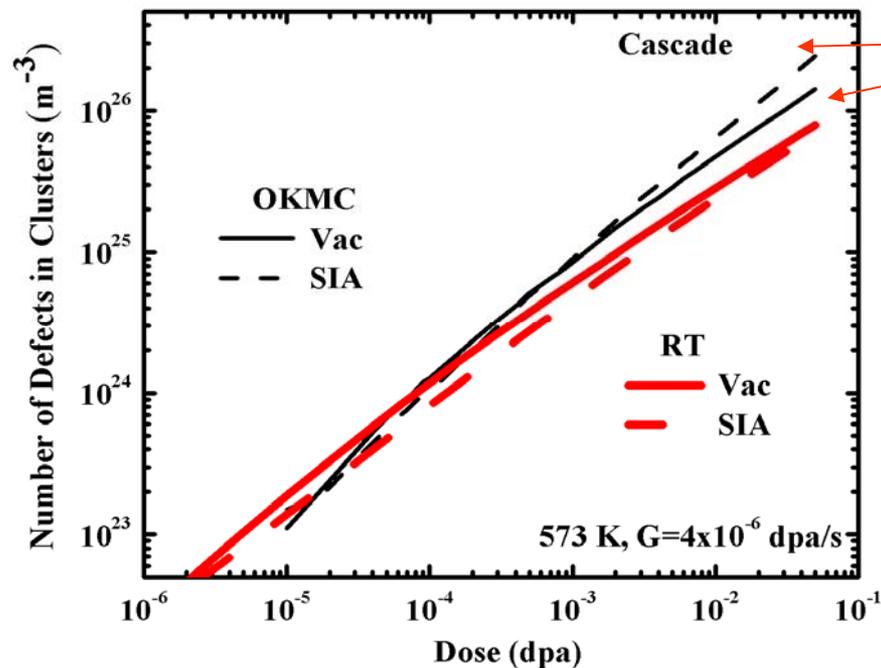
$T > 200^\circ\text{C}$  significant fluctuations in  $C_v$ , about 300 vacancies in OkMC box at  $200^\circ\text{C}$

$T > 150^\circ\text{C}$  significant fluctuations in  $N_{\text{vcl}}$ , about 50 clusters in OkMC box at  $150^\circ\text{C}$ ,  $< 10$  at  $200^\circ\text{C}$

one defect in box



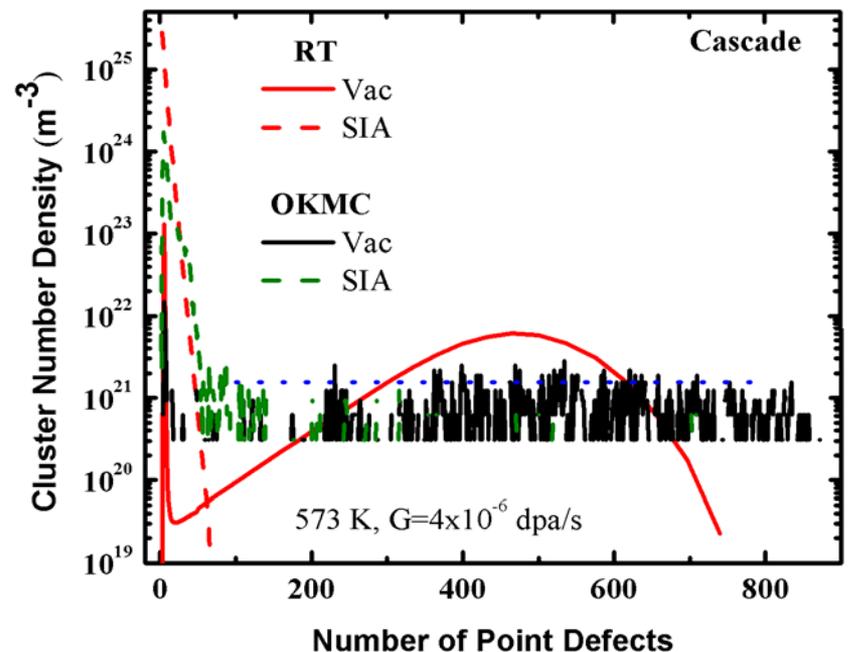
# Effect of Cascade production: 200°C



difference in behavior of defect accumulation

number of interstitials accumulated in SIA clusters should approach the number of vacancies in voids, slight excess in vacancy accumulation due to the dislocation-interstitial bias

spurious cluster nucleation due to non-physical fluctuations leads to excess large clusters, particularly for SIA



# Summary: RT-OkMC Comparison

- Common issues, primarily related to parameterization and basic model formulation
  - e.g. binding and migration energies for small point defect clusters, nature of defect migration (1-D vs 3D)
  - are all (or all relevant) mechanisms and reactions accounted for?
- Both approaches produce similar results on well-posed problems, similar dependence on key irradiation and material variables
- Issues for RT
  - account for effects of discrete time and spatial events, e.g. primary damage
- Issues for OkMC: primarily computational
  - limited to relatively low doses
  - system size limitation constrains available regime of irradiation temperature and dose rate
- RT-OkMC differences remaining
  - cluster densities when statistical nucleation difficult
  - damage accumulation under cascade damage conditions
  - uncorrelated vs correlated cascade debris

## caveats on mesoscale modeling

- *ab initio* methods and MD can provide improved estimates of material parameters, e.g. point defect formation energies, primary radiation damage parameters
  - former is largely limited to simple materials and small atomic systems, providing limited information on diffusion and defect formation energies, and latter is limited by range of materials for which adequate interatomic potentials can be developed
- most importantly, keep in mind that models are inherently incomplete, sometimes we know what we don't know, sometimes we don't, e.g.:
  - solute effects (alloy thermodynamics) not accounted for in most rate theory models
  - real materials are not spatially homogeneous (see examples above)
  - from MD: neutron irradiation produces small point defect clusters as well as the monomers, ~10-60% defects in clusters, vacancy and interstitial clustering fractions different
  - from MD: observed diffusion behavior more complex than simple 3D, small clusters also mobile, alternate diffusion mechanisms change reaction kinetics (sink strengths of extended defects)

# Overall Summary

Substantial progress in understanding and predicting the behavior of materials has been provided by theory and computational modeling

Further progress requires additional research in the following areas:

- Electronic structure calculations to obtain intrinsic and defect properties in iron and its alloys, including the effects of He and H
- Computationally-efficient, physically-robust interatomic potentials for multicomponent alloys, including effects of directional bonding and magnetism
- Advanced atomistic and mesoscopic models describing the many defects and processes that interact in complex ways in multicomponent, multiphase materials
- “Properly” linked and multiscale (atomistic, mesoscopic, and continuum) deformation and fracture models for predicting hardening, plastic instability, changes in ductile-to-brittle fracture, dimensional instability, and creep/creep rupture behavior under realistic loading conditions