Atomic-scale Simulation of Radiation Damage in Structural Materials

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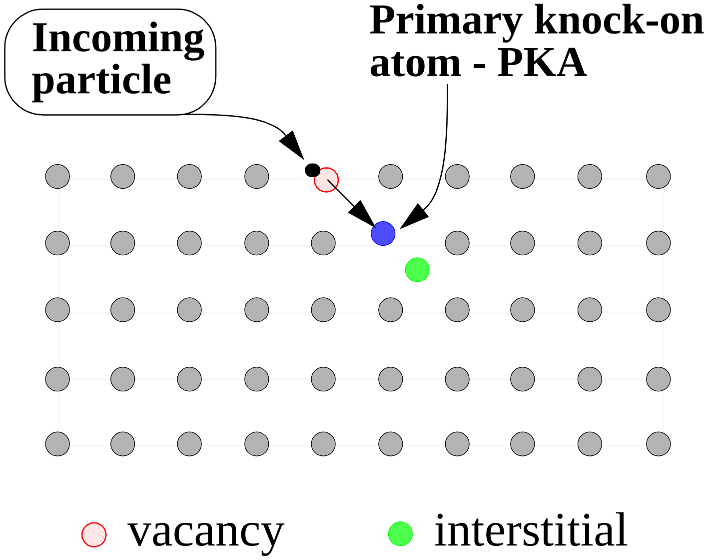
Why do we care about radiation damage in structural materials - What is the impact?

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

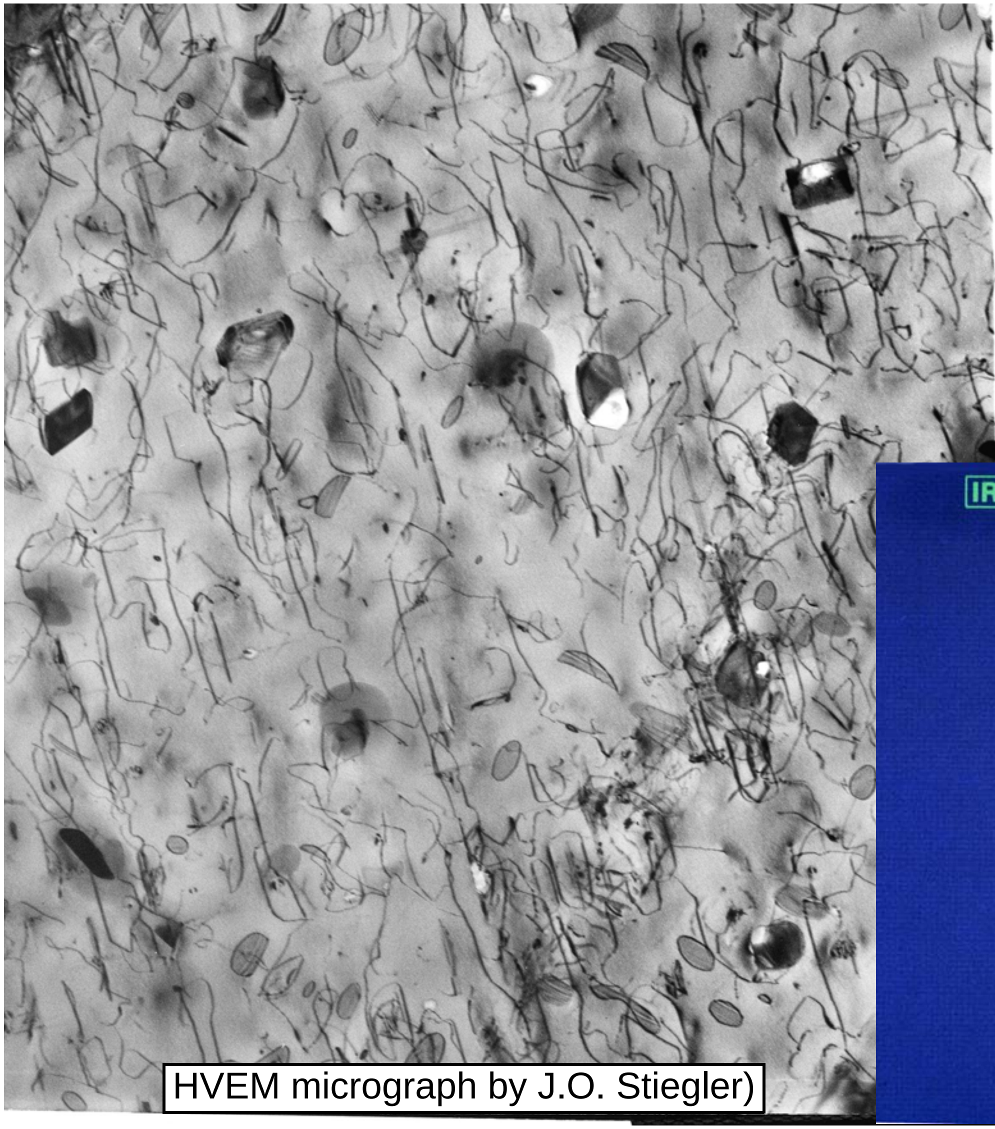
- grain size, other internal interfaces
- dislocation density
- size and density of second phase precipitates

Irradiation with energetic particles leads to atomic displacements

- neutron exposure can expressed as particle fluence (#/m^2) or a dose unit that accounts for atomic displacements per atom - dpa
- lifetime component exposures are in the range of ~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



HVEM micrograph by J.O. Stiegler)

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

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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

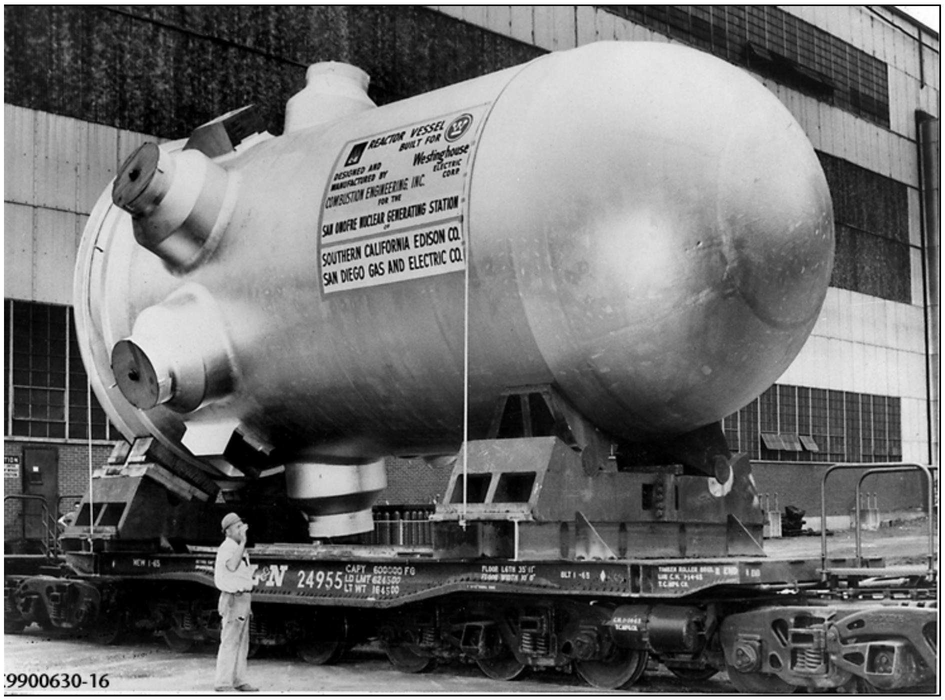
- costs for design and execution of reactor irradiations
- costs of post-irradiation examination of radioactive materials
- declining facilities for both irradiation and examination
- combinatorial problem: broad range of materials, phenomena, and irradiation conditions - coolants, temperature, loading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

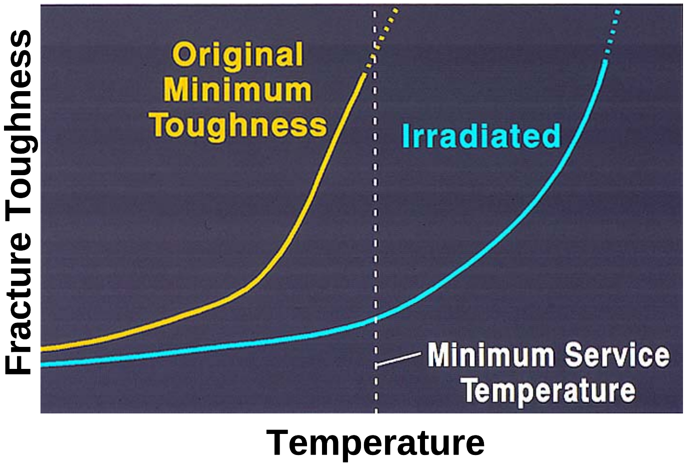
Recent advances in computational hardware, computational science, ... make it more feasible than ever to aggressively attack this challenge

- 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

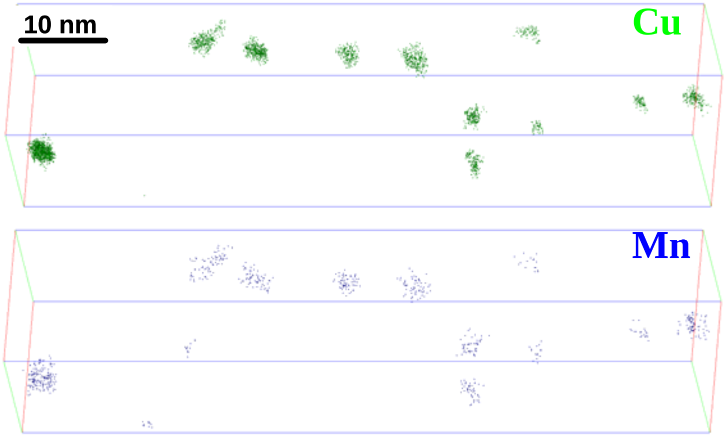
Understanding the Effects of Irradiation on Structural Materials Requires Multiscale Modeling and Experiments

- 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

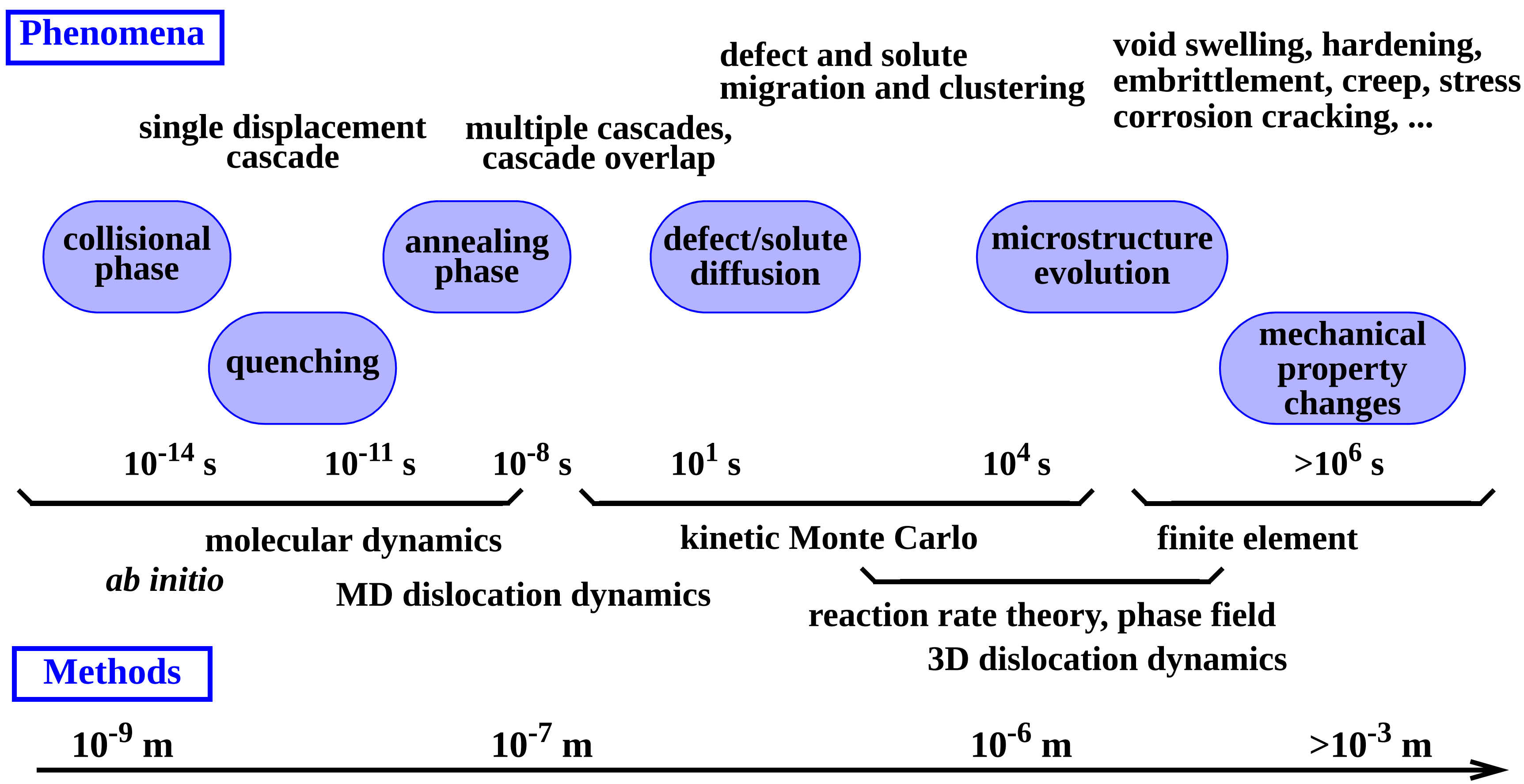




- copper (and Ni, Mn, ...) -enriched clusters in neutron irradiated model RPV steel, APFIM data: Miller, ORNL



Schematic diagram: relevant phenomena and computational methods



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

Provide brief illustrations from different examples of radiation-induced phenomena and property changes, emphasize models and mechanisms

- primary damage formation, molecular dynamics and kinetic Monte Carlo
  - evolution from fast fluence (E>1.0 MeV) to dpa, energy spectrum effects
- ab initio (VASP), accounting for He defects in iron, influence on defect properties and interatomic potential
- atomistic simulation of dislocation-defect interactions, molecular dynamics
- mesoscale models of microstructural evolution
  - reaction rate theory, microstructural evolution, illustrate loose (parameter passing) multiscale modeling
  - compare reaction rate theory and object kinetic Monte Carlo
  - displacement rate (neutron flux) effects in RPV steels, kinetic embrittlement model

Aspects of Primary Radiation Damage Source Term

I. 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 ~ 80-100 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 up to ~20 MeV, peak at ~0.65 MeV, ratio ϕ(peak)/ϕ(10 MeV)~350
  - DT fusion at 14.1 MeV
  - displacement cross section minimum at ~1 keV (elastic scattering limit) for iron
- thermal neutrons
  - typically E<0.5 eV, kT_{room} =0.025eV)
  - produce low energy recoils from (n,γ) 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,γ) reactions
  - generate low energy recoils (similar to thermal neutrons) by elastic scattering

< Note: displacements from either thermal neutrons or electrons can be significant in certain cases, e.g. HFIR RPV (e^-), heavy-water (HFBR, Halden) or graphite moderated (MAGNOX) cores >

- nuclear transmutation products
  - gases: primarily hydrogen and helium from (n,p) and (n,α) reactions
  - solid: (n,p), (n,α), (n,2n), (n,γ) with subsequent β 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 ϕ_{th}=2.5x10^{26} n/m^2 (~6 months in HFIR) converts 1% of Al to Si

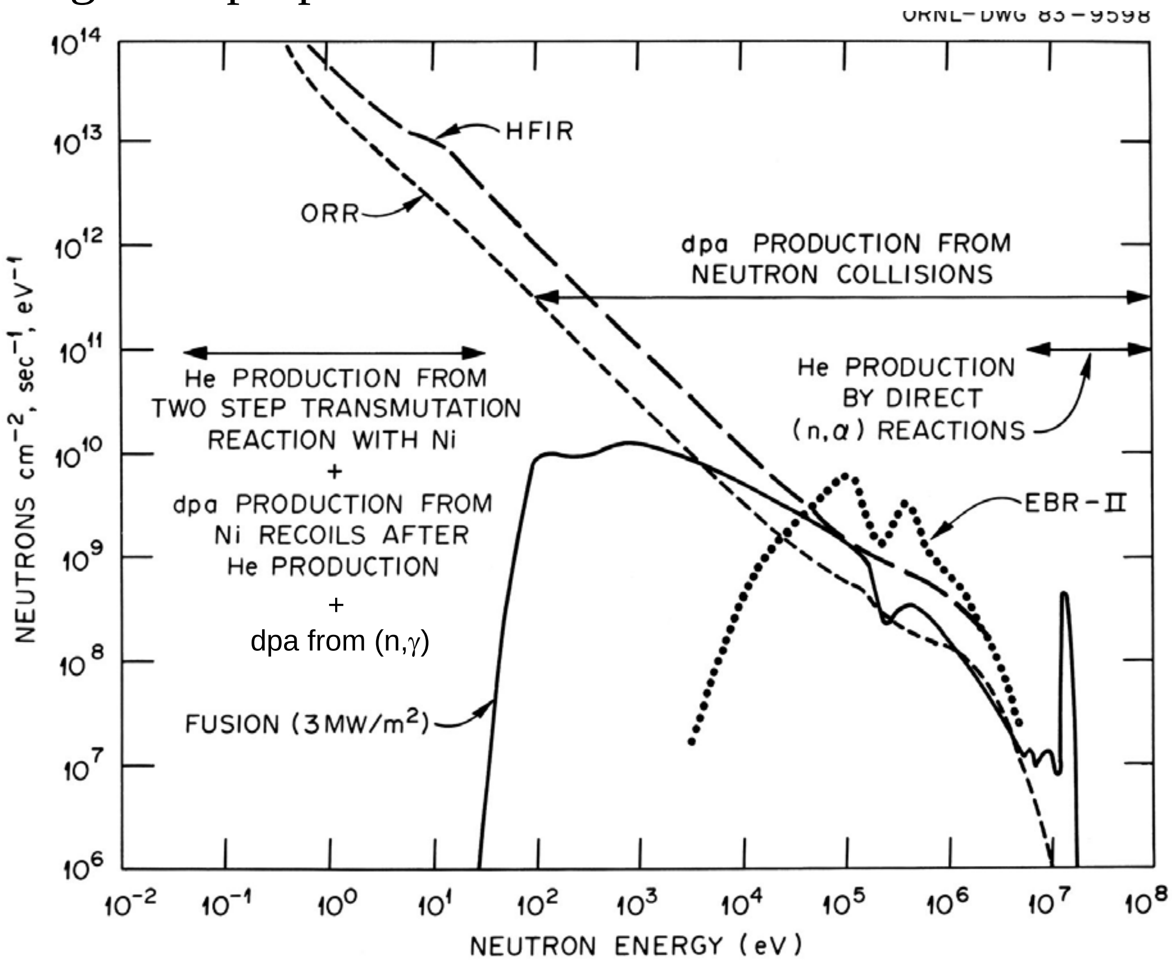
Primary Radiation Damage, con't.

II. 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
  - significant need/opportunity for dosimetry experiments and calculations
- proposed D-T fusion materials irraadiation facility
  - 40 MeV D^+, 125 mA (x2) on liquid Li, D-Li stripping reaction, 14 MeV+ neutrons

Significance of neutron energy spectrum, relevance of dosimetry

- different damage mechanisms relevant for different particle energies
- absolute damage rate proportional to neutron flux level



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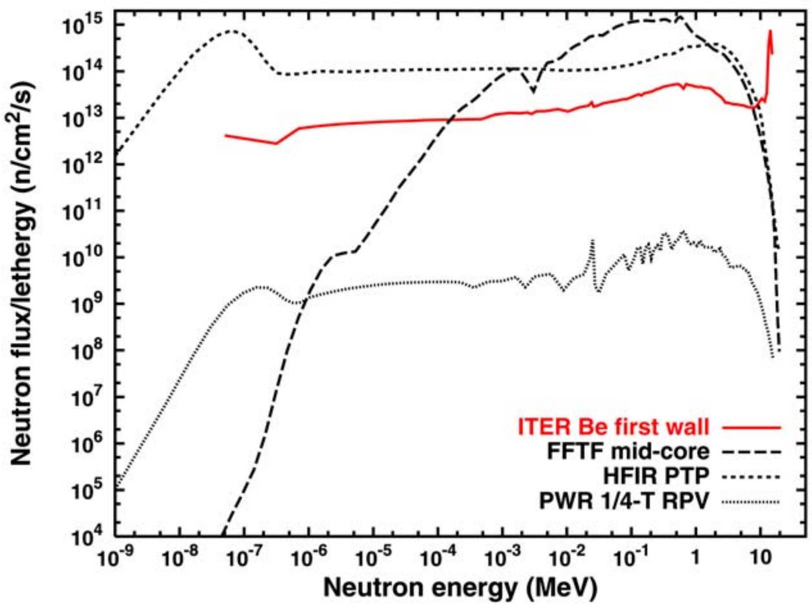
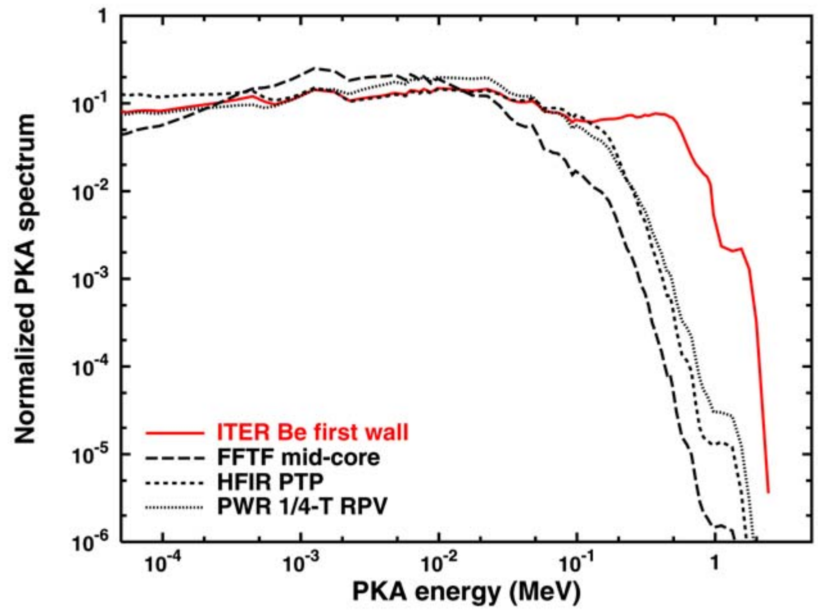
Comparison of representative neutron and corresponding PKA energy spectra





- 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

(1) Molecular dynamics simulation of primary damage

MD simulations provide opportunity to use improved interatomic potentials to investigate displacement cascade evolution, e.g. effects of crystalline lattice

Classical MD, typical implementation:

- 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_{dam}= kinetic energy lost in elastic collisions,

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

Range of interatomic potentials 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 NRT model for number of defects produced by an atom following a collision with a high energy neutron

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

10 keV MD example

Partial Fe Cascade Database at 100K

MD Cascade Energy (keV)	Corresponding neutron (MeV) and PKA (keV) energies		NRT displacements	Number of Simulations	Typical simulation cell size (atoms)
0.1	0.00335	0.116	1	40	3,456
0.2	0.00682	0.236	2	32	6,750
0.5	0.0175	0.605	5	20	16k/54k
1.0	0.0358	1.24	10	12	54k
2.0	0.0732	2.54	20	10	54k
5.0	0.191	6.6	50	9	128k
10.	0.397	13.7	100	15	125k/250k
20.	0.832	28.8	200	10	250k
50.	2.28	78.7	500	9	2.249M
100.	5.09	175.8	1000	10	5.030M
200.	12.3	425.5	2000	9	16M

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

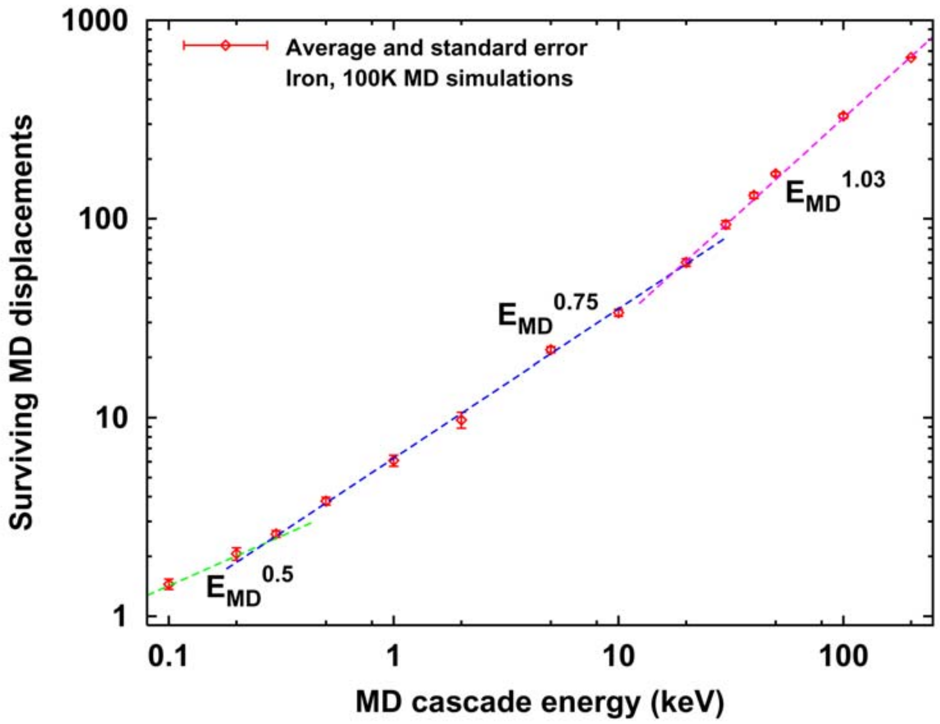
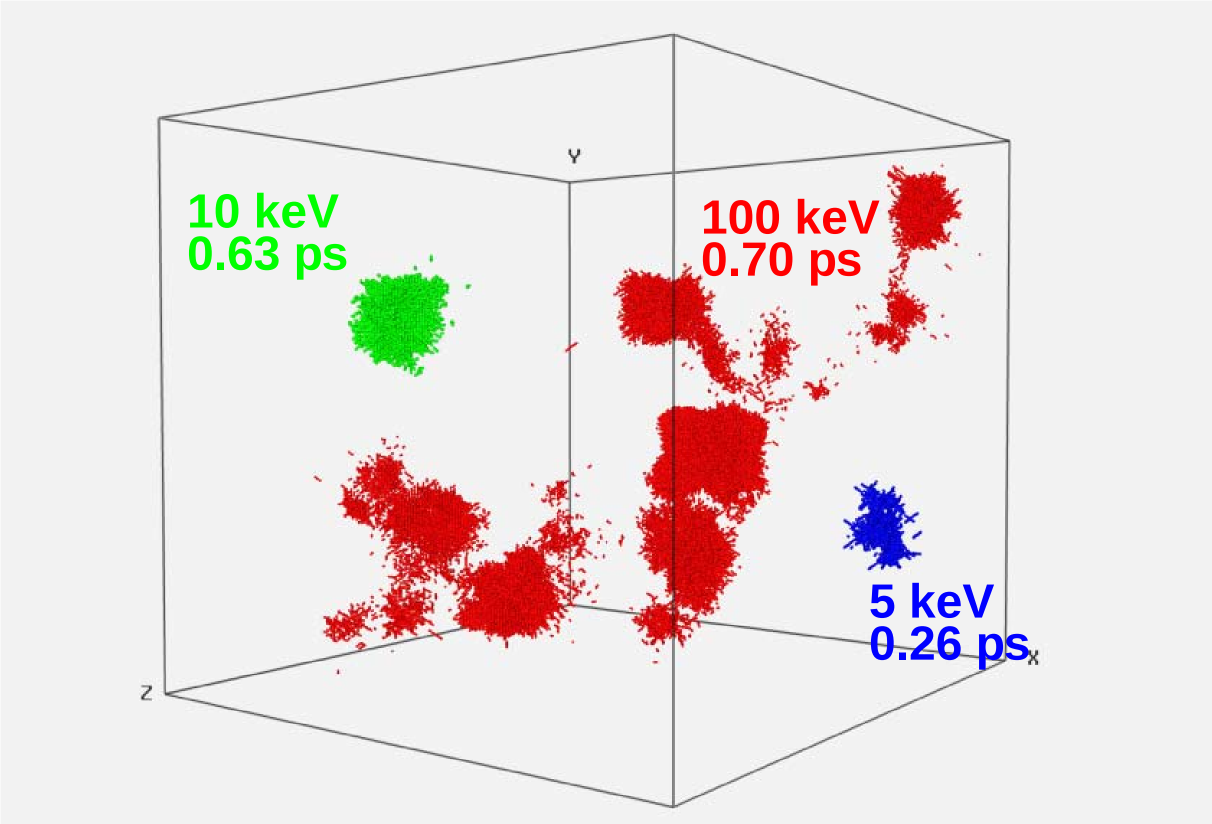


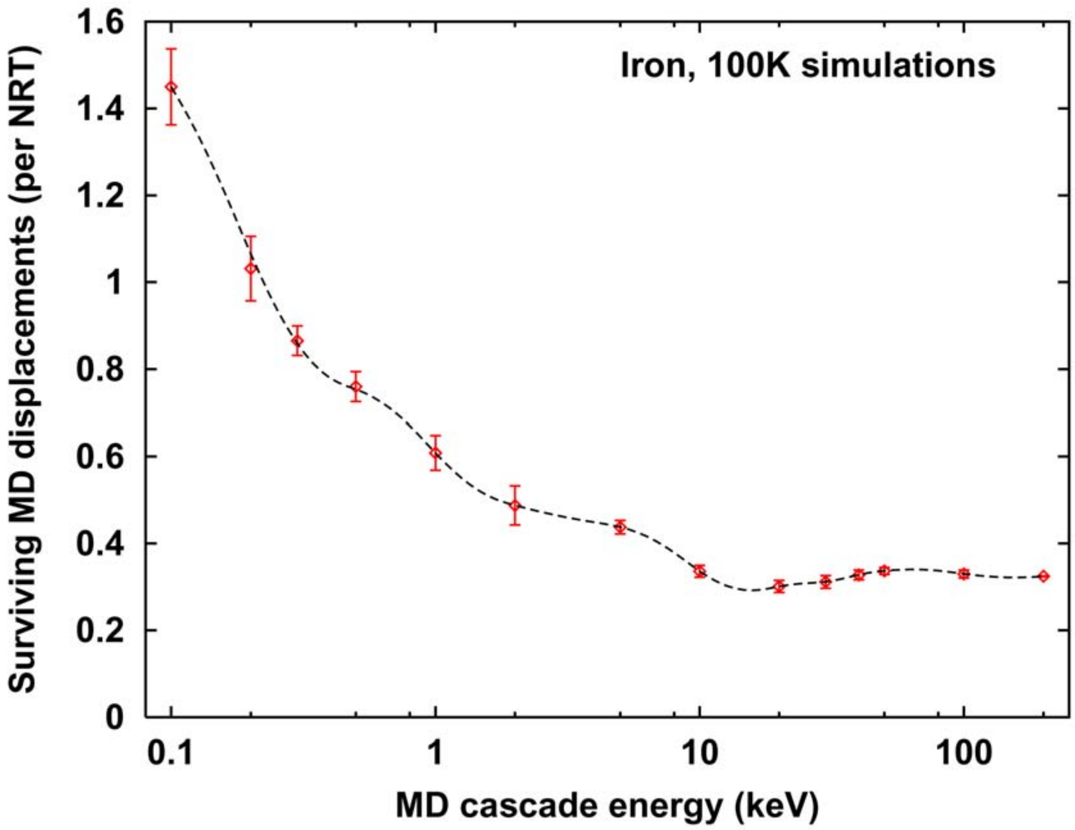
Illustration of subcascade structure at peak damage condition for 5, 10, and 100 keV cascades at 100K



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

Changes in damage production efficiency best illustrated by normalization to NRT displacements

Total surviving defects



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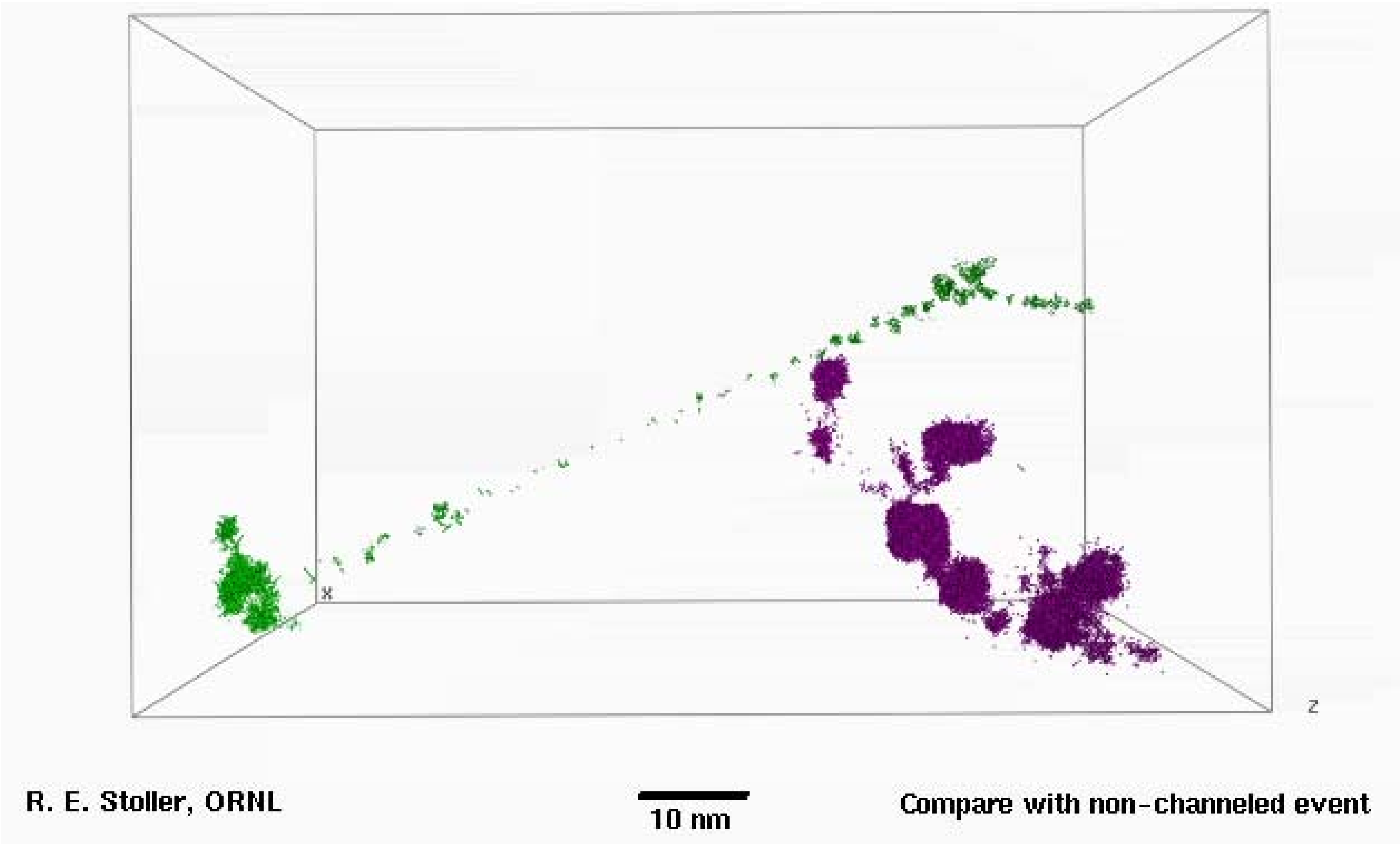
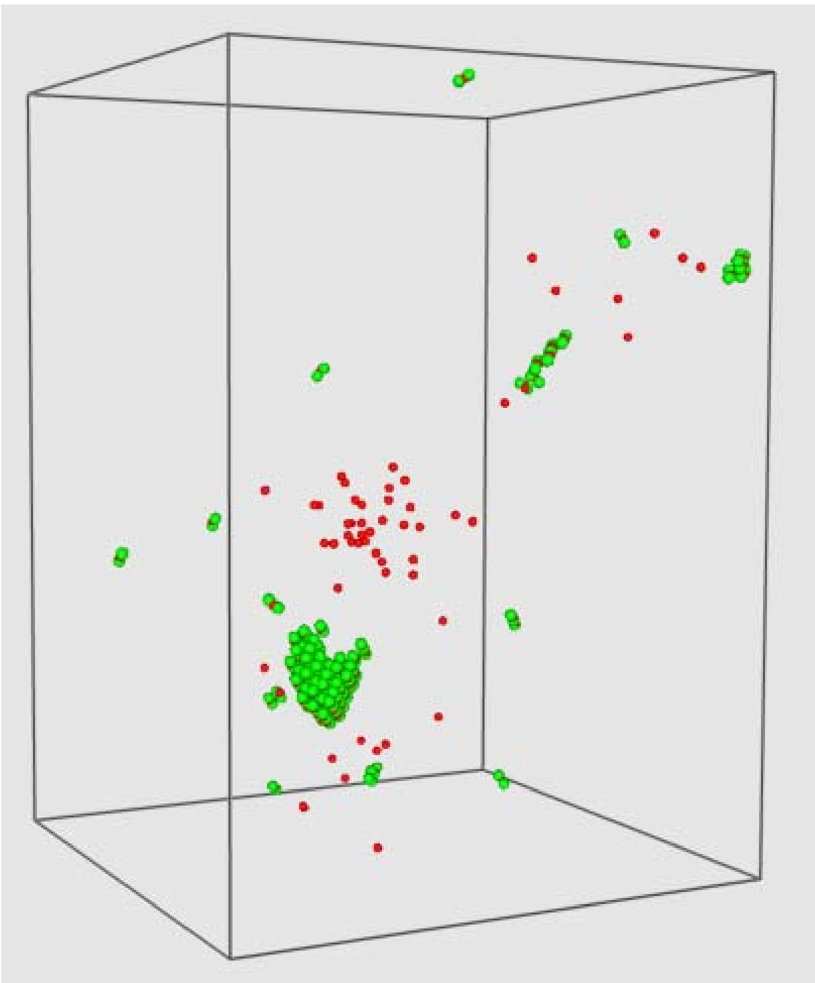
Low angle (low energy transfer) collisions lead to channeling at high cascade energies

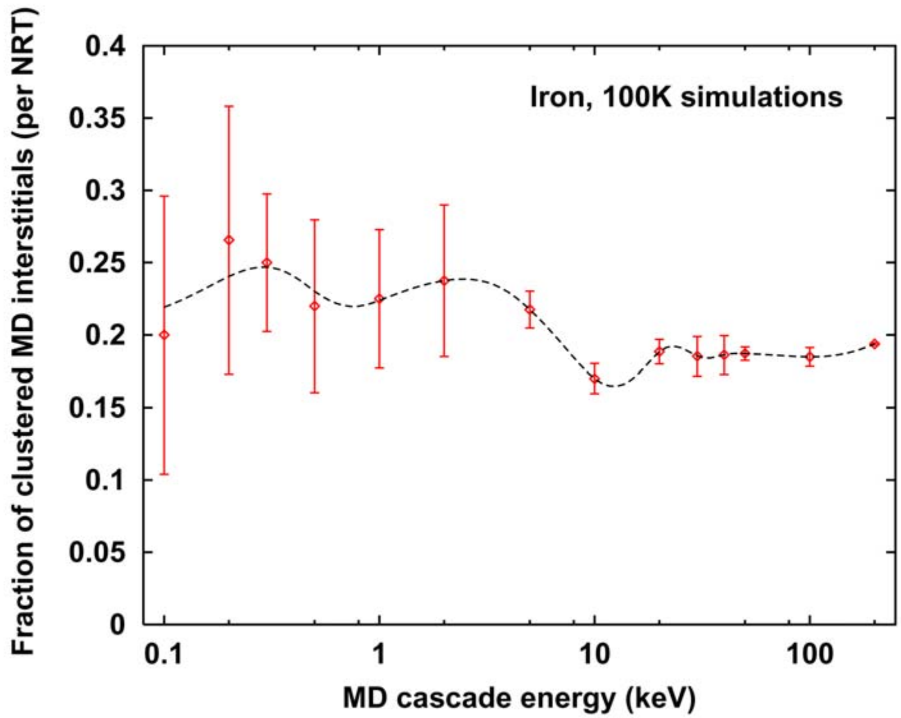


- low energy "cascades" between subcascades have higher damage production efficiency - leads to slight increase in survival fraction above ~20keV

Many surviving defects are in clusters formed during cascade event

- 20 keV cascade, 600K





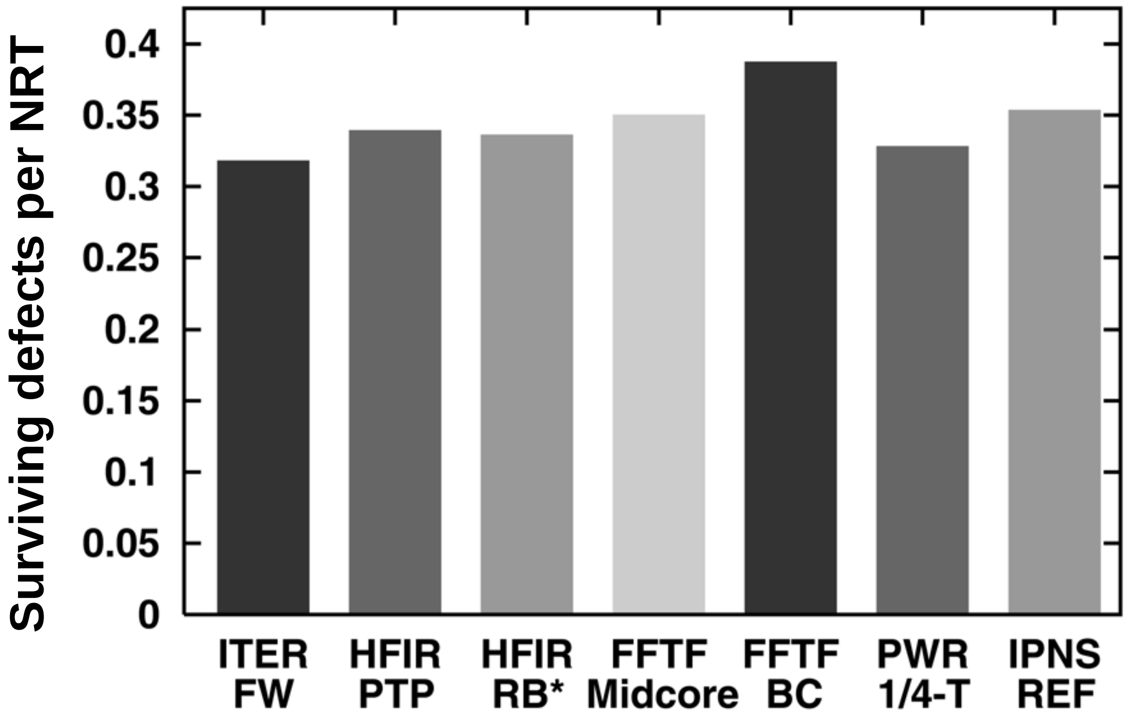
- incascade clustering significant for heterogeneous nucleation of extended defects

Evaluation of neutron energy spectrum effects using MD results and calculations from SPECOMP and SPECTER

- Energy-dependent functions were fit to MD defect production results
- MD-based functions were used as a factor multiplying the standard displacement cross section equations as a function of the damage energy, T_{dam} in SPECOMP to produce surviving defect and clustered interstitial cross sections on a 100 point neutron energy grid.
- cross sections were added to the SPECTER libraries to produce spectrum-averaged values for the point defect and interstitial clustering fractions for various neutron spectra

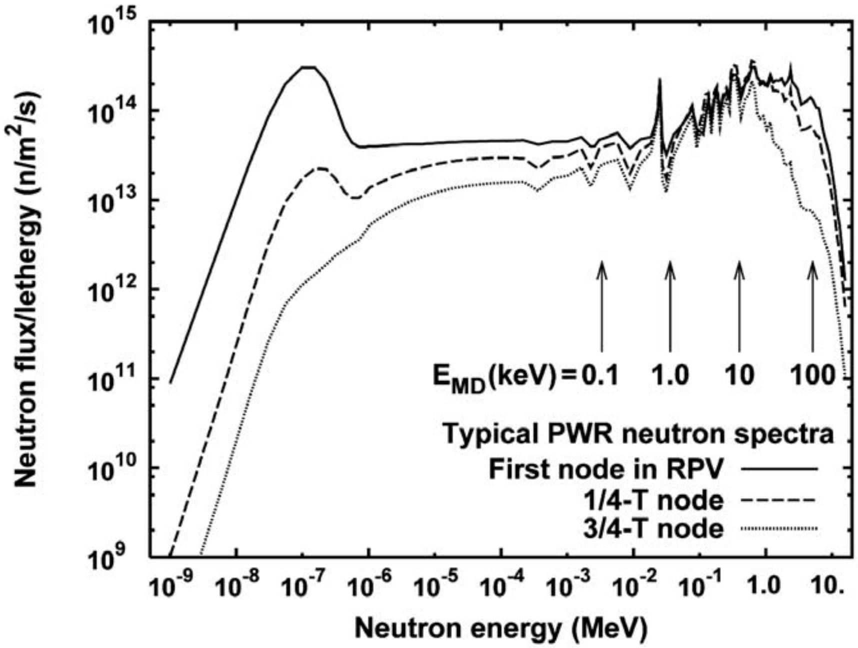
* Larry Greenwood, PNNL

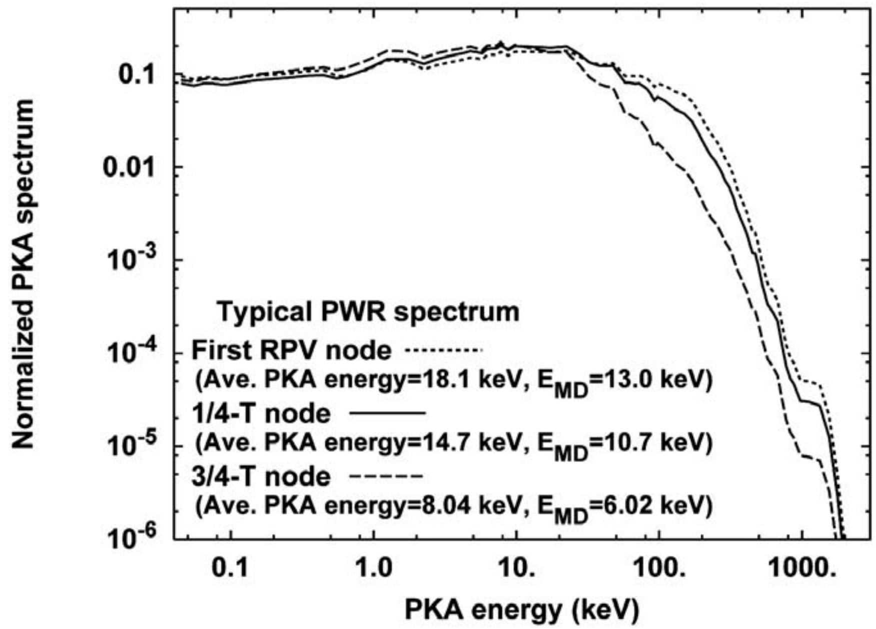
Spectrum-averaged total defect survival



- spectrum sensitivity not strong due to weighting of defect survival energy dependence with that of PKA spectrum
- similar results for point defect clustering fraction, stronger influence on cluster size distribution

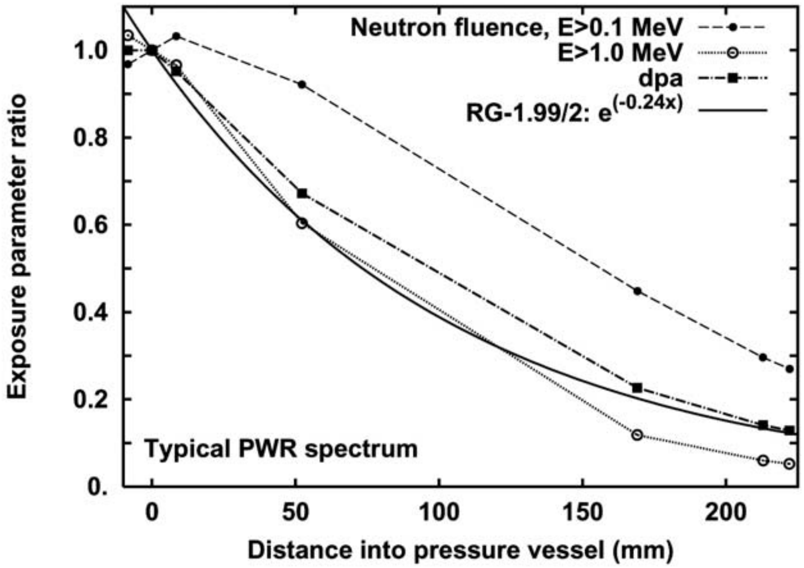
Energy spectrum effects: Through-RPV attenuation

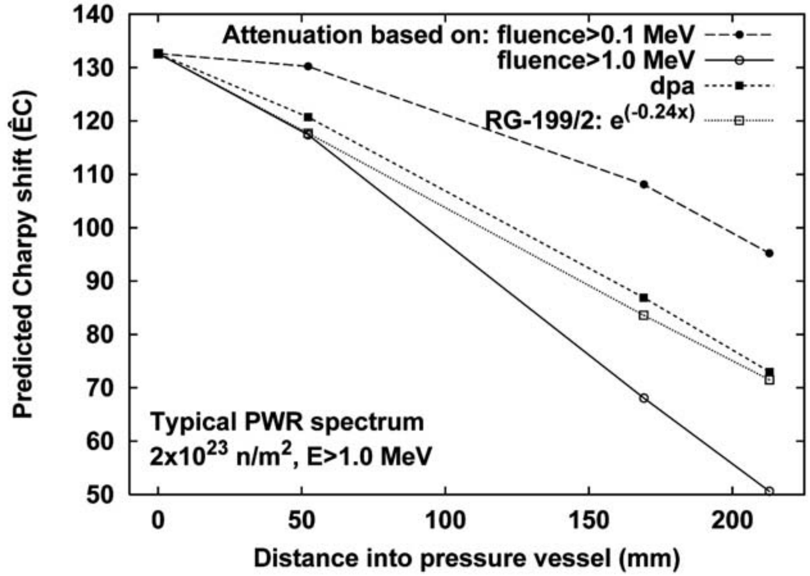




- spectrum reduced at high and low energies
- corresponding changes in PKA spectrum

Damage attenuation:
Comparison of alternate exposure or correlation parameters





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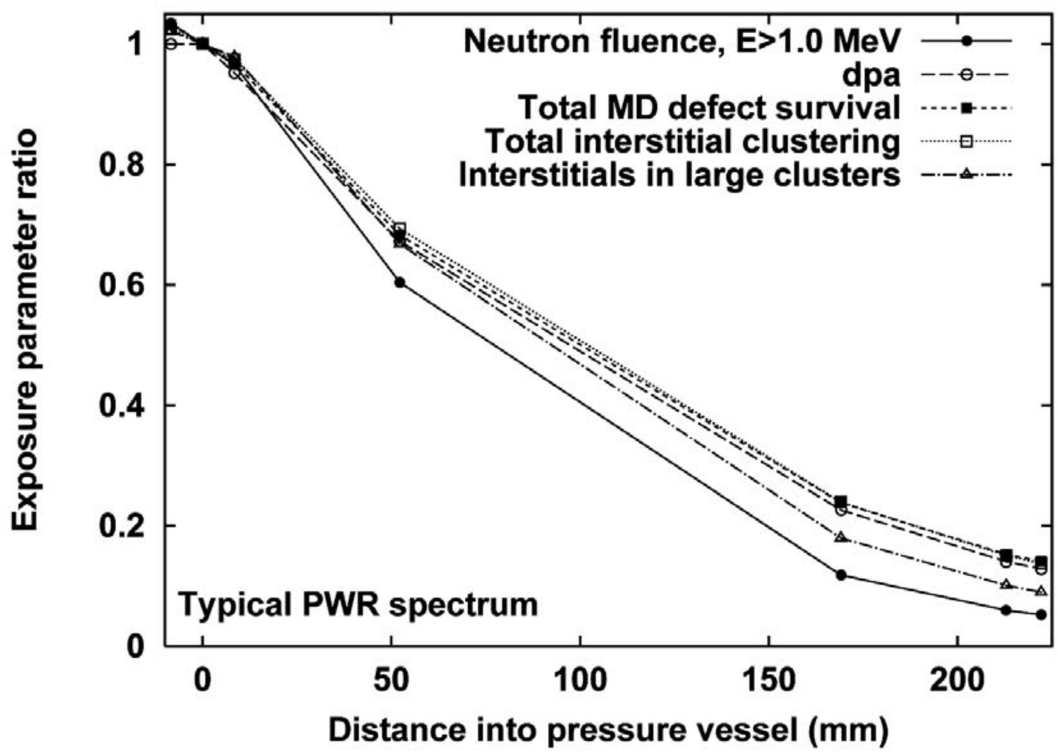
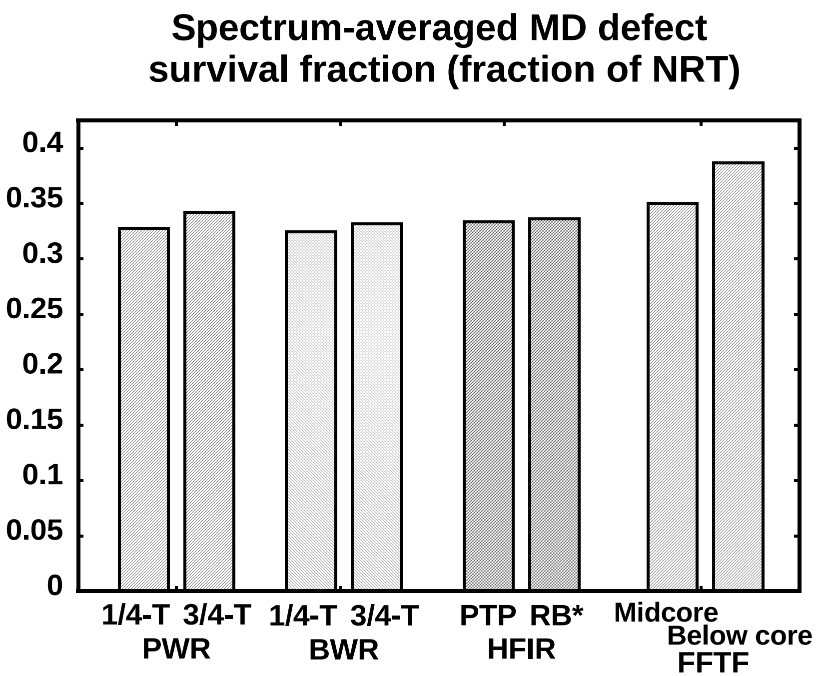
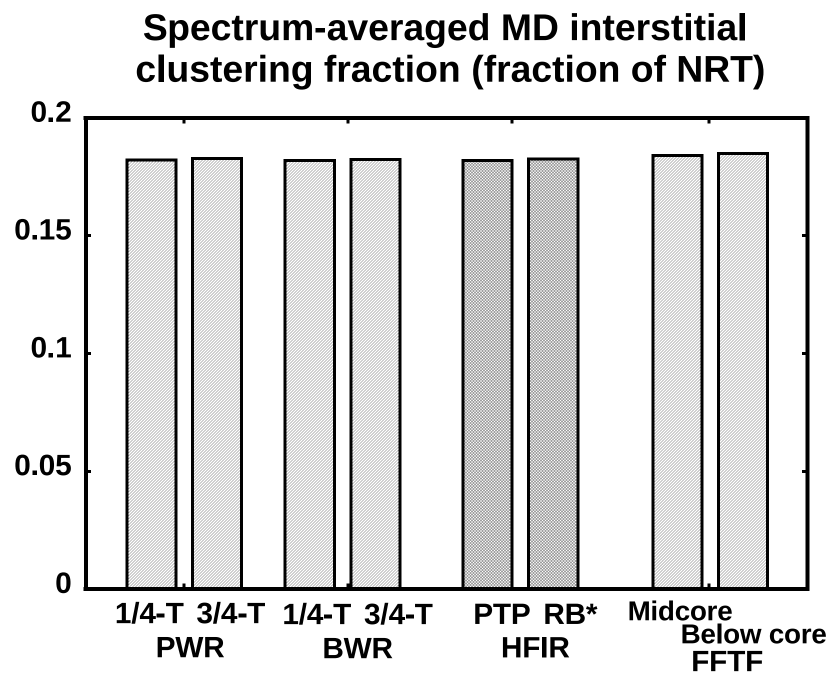
Spectrum-averaged MD parameters not greatly different from dpa



Spectrum-averaged MD parameters do not show significant difference between BWR and PWR spectra





Summary: Primary Radiation Damage Simulation

- extensive MD cascade studies have been carried out, largest database for Finnis-Sinclarium “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
- atomistic simulations support coarser scale models, provide opportunity for practical investigation of neutron (PKA) energy spectrum effects,

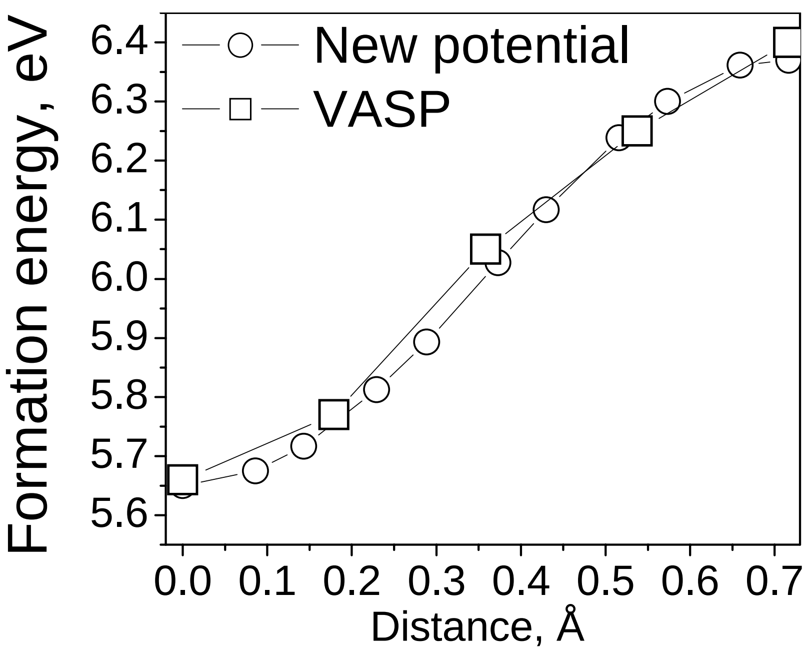
(2) Development of improved Fe-He interatomic potential based on *ab initio* calculations

Motivation is impact of helium is produced in neutron-irradiated materials by (n, α) transmutation reactions

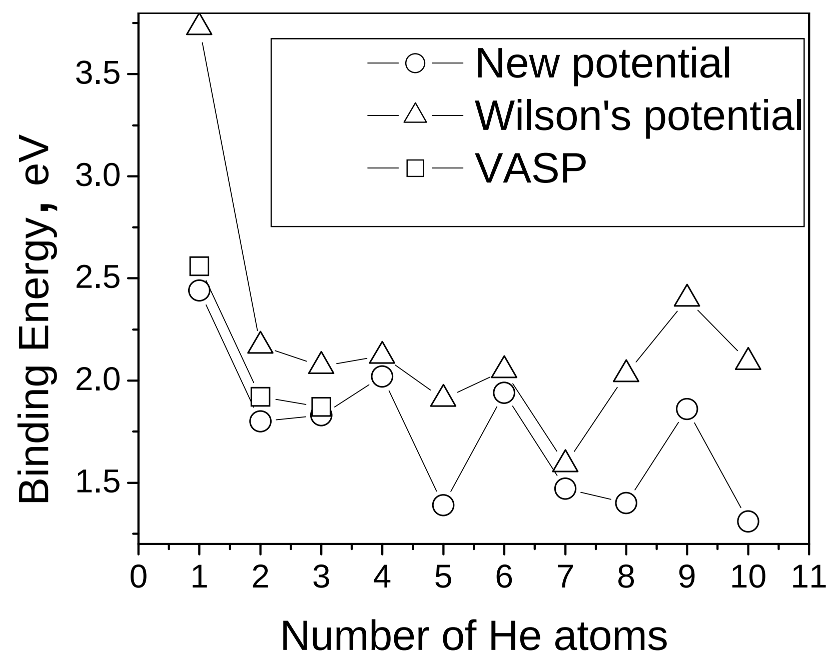
- experimentally observed to alter radiation-induced microstructural evolution, e.g. enhance swelling by stabilizing bubbles and cause embrittlement by segregating to grain boundaries
- DFT calculations (VASP) used to compute properties single He defect and small He clusters in iron
  - plane-wave basis set using PAW pseudopotentials, generalized gradient approximation
  - plane-wave energy cutoff was set to 300 eV
  - 54 and 128 atom supercell using 10-75 k-points
- Fe-He pair potential fitted to VASP forces reproduced relaxation energies, but strongly over-estimated defect formation energies
- many-bodied empirical potential Fe-He obtained by fitting the DFT results, initially used Finnis-Sinclair iron

- in contrast to earlier work tetrahedral site found to be the preferred interstitial site, unexpected hybridization of Fe-He electron orbitals and small affect on Fe magnetic moment
- use of many-body He-Fe potential based on Finnis-Sinclair Fe shown
- He-vacancy cluster binding energies found to be substantially different than earlier work

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



binding energy of helium to single vacancy



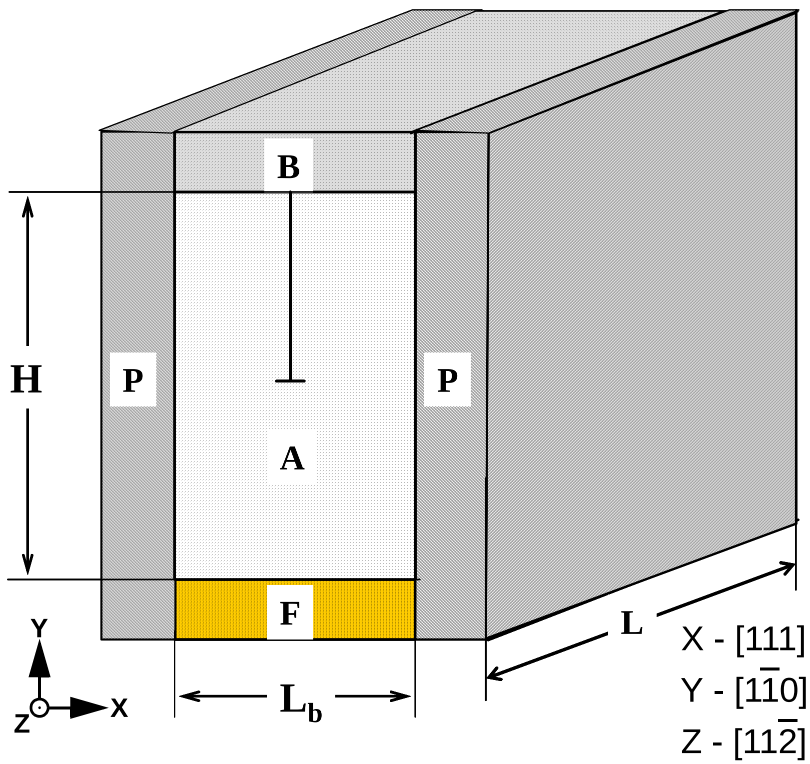
ab intio applications in radiation damage modeling

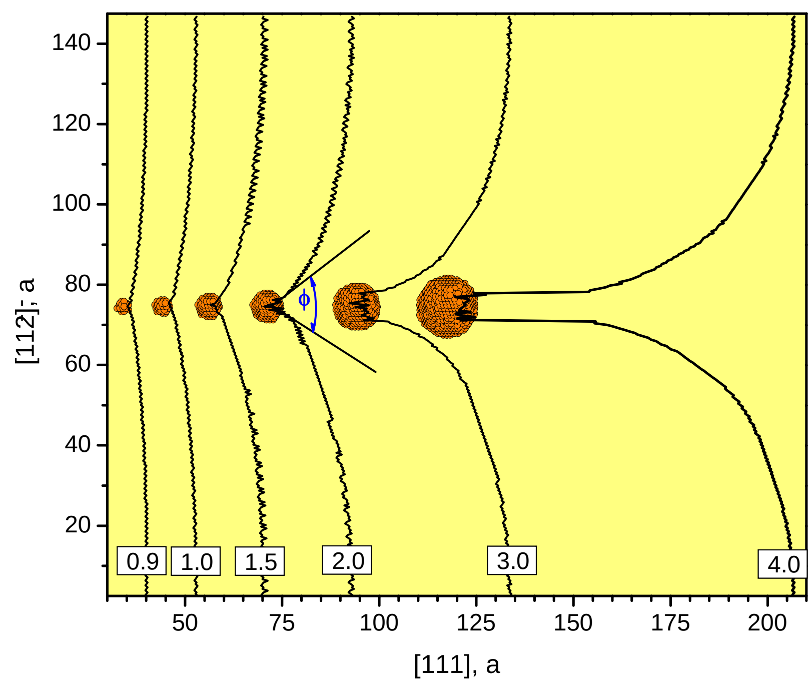
- support development of improved interatomic potentials for atomistic simulation
- He-Fe one example
  - theory-based EOS for He in Fe, for application in mesoscale and continuum models
  - MD-based investigation of He-vacancy defects
  - use MD simulations to provide parameters for cluster dynamics models to investigate evolution of He-vacancy clusters leading to bubble nucleation
- scaling up of *ab initio* calculations to examine larger defects
- similar work needed for other materials
  - pure metals and alloys
  - ceramics
  - nuclear fuel, theory development required for actinides, 5f electron system
- effects of hydrogen need similar investigation

(3) 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

(a) Atomistic nature of dislocation-defect interactions

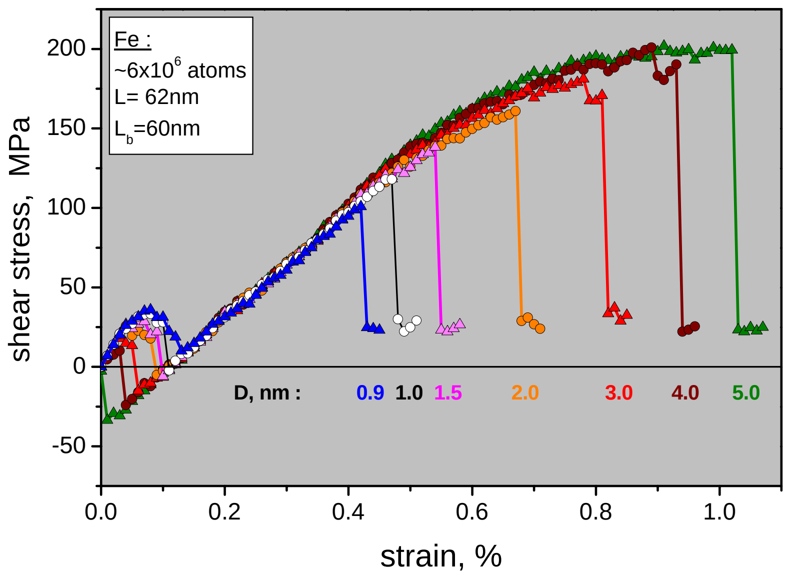
- radiation-induced increase in yield strength based on simple hardening theory for dispersed barriers

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

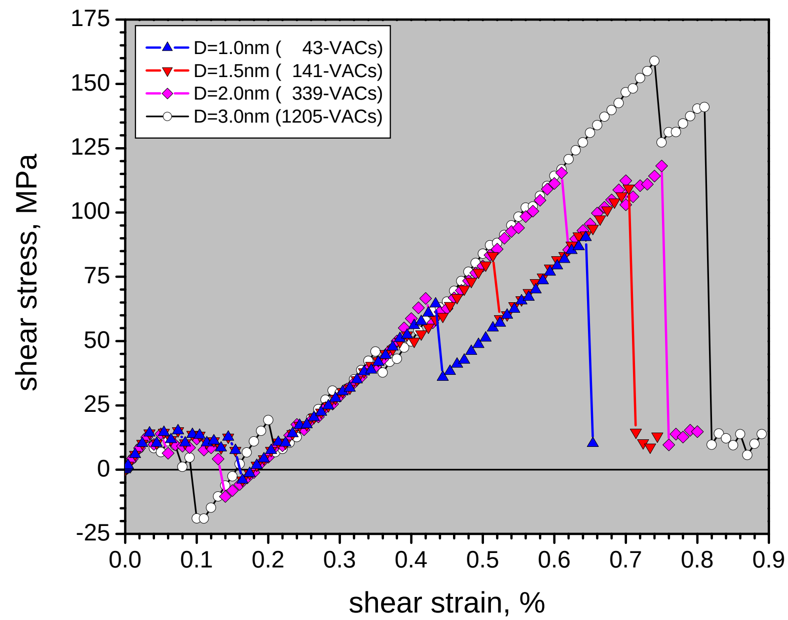
where T is the Taylor factor (3.06), N and d are the radiation-induced defect density and mean diameter, and α is the so-called barrier strength

- 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.

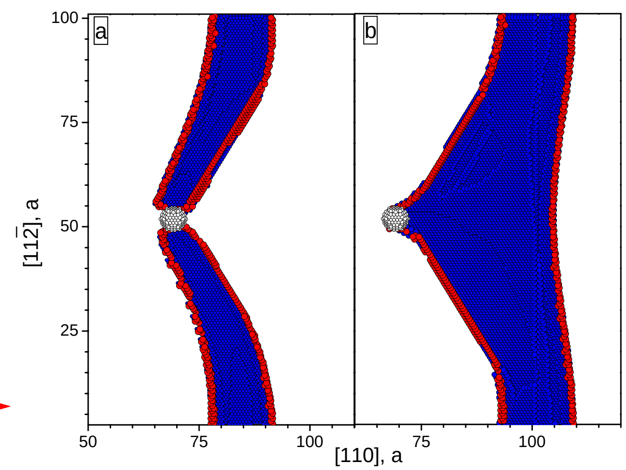
- elasticity-based dislocation models can not account for atomistic details of dislocation-defect interactions



voids in iron



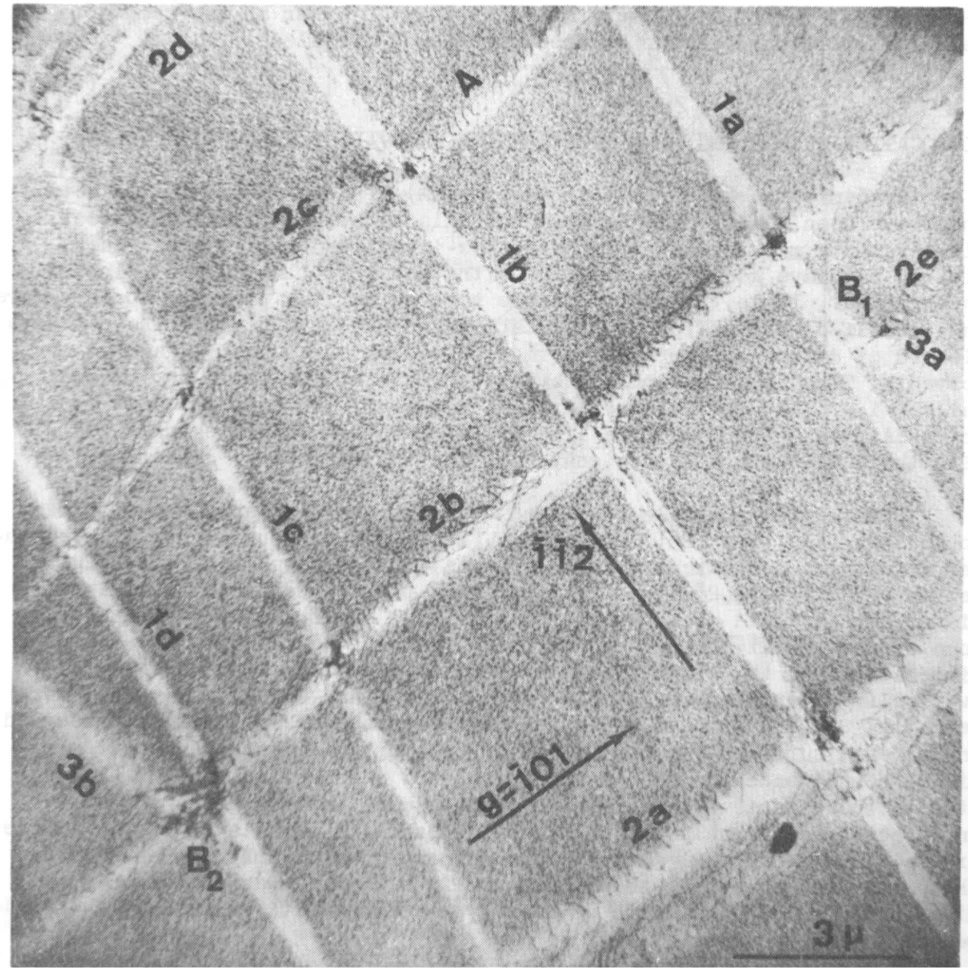
voids in copper



dissociated dislocation in copper

(b) 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 4.4×10^{22} n/m^2 at ~50°C and strained to 6% at room temperature



MD Example: 12nm (820 vac) SFT FS

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
- 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
- simulations with improved potentials for alloys, and materials such as ODS steels

(4) 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 reaction rate theory modeling

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

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

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

where the ∇ 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:

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

- 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:

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

  - 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 - <u>smallest</u> 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

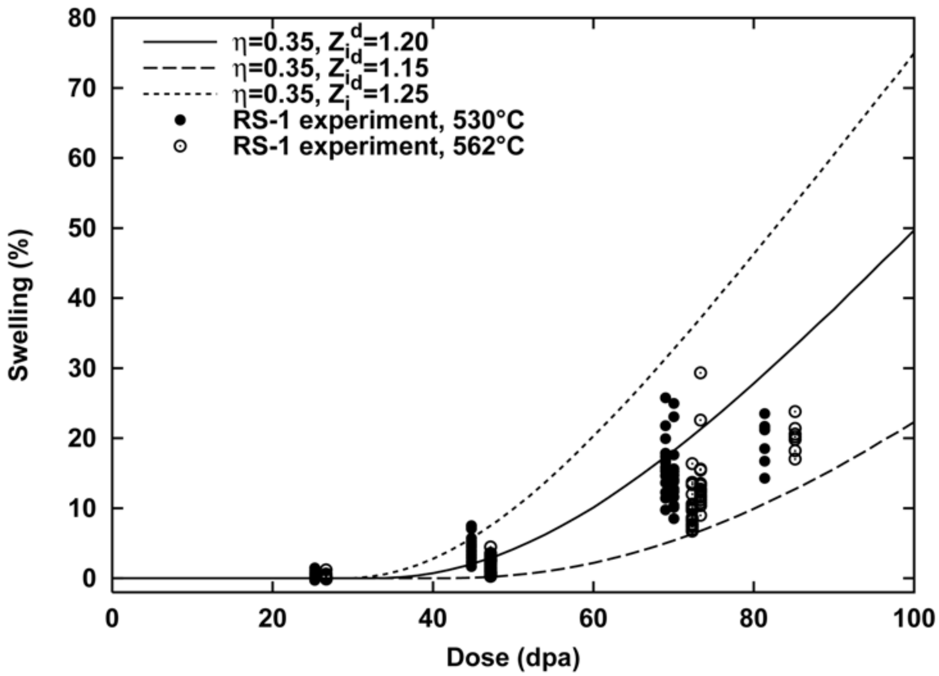
- rate equations can be written to describe the evolution of extended defects, void growth rate and void sink strength:

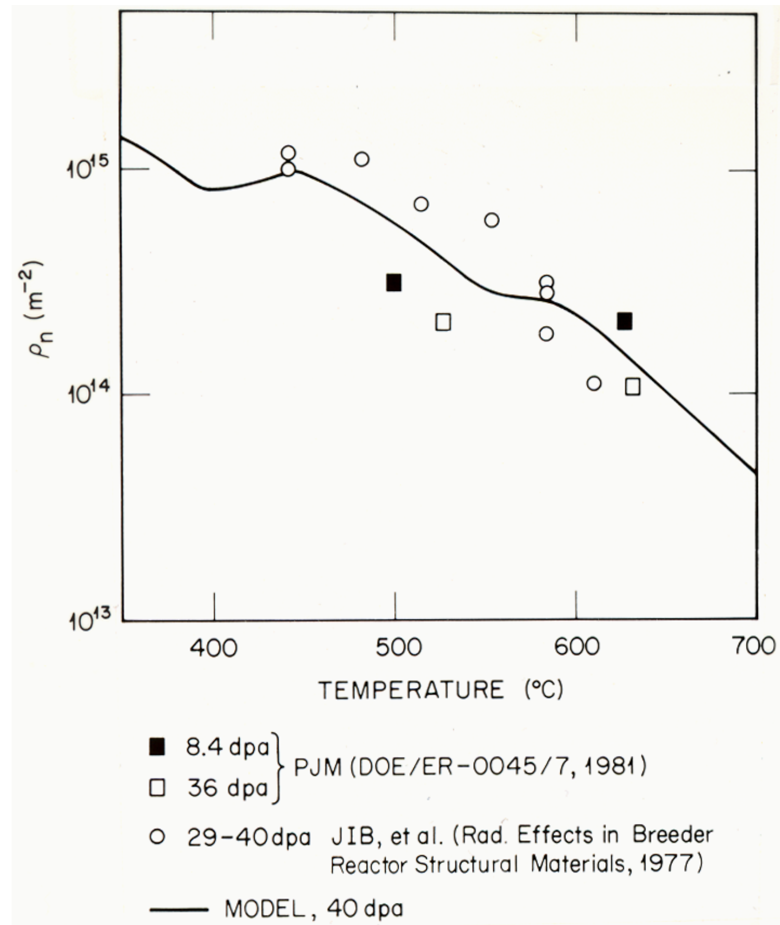
$$S_{i,v}^{v} = Z_{i,v}^{v} 4\pi r N_v \left(1. + r_v \left(S_{i,v}^{T}\right)^{0.5}\right)$$

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

- similar 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





R. E. Stoller, ORNL

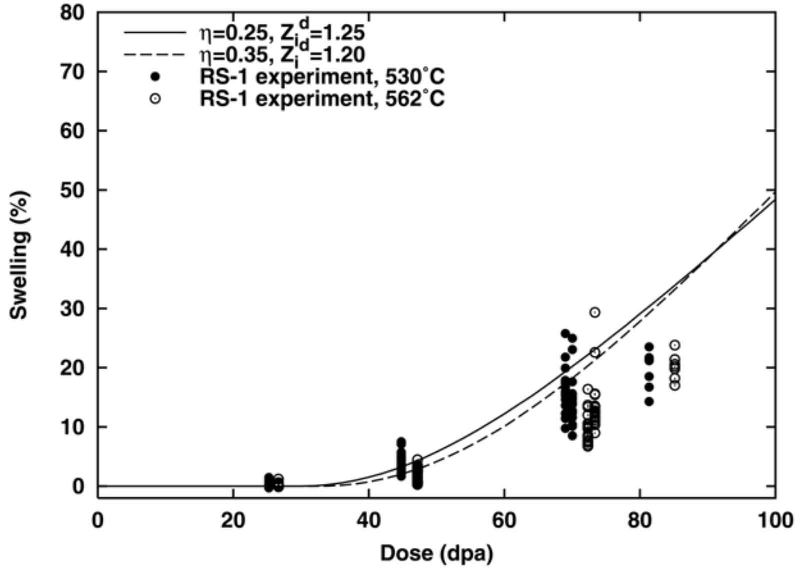
However, the success of these models in fitting data can be deceiving (the devil is in the details ...)

- 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 Sd, swelling rate is proportional to product of h and net dislocation bias (Zid -1):

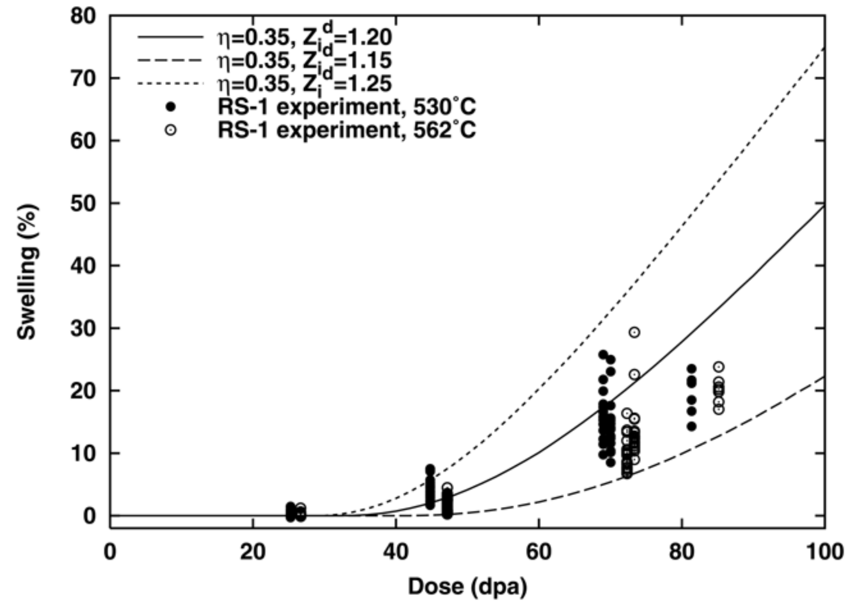
$$\frac{dV}{dt} = \frac{\eta G_{dpa}}{S_v^d}\left(Z_i^d - 1\right)$$

- in this case, data fitting can not be used to obtain unique set of model parameters.
- 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, provide limited information on diffusion and defect formation energies, and latter is limited by range of materials for which adequate interatomic exist

- use of MD-based damage parameters and calculated PKA spectrum for a given irradiation environment (here the FFTF) can be used to obtain spectrum-averaged η, provide estimate for bias



- arbitrary Z_i^d and η



- η 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

- more generally, need to keep in mind that such 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)

Comparison of kinetic models: Reaction Rate Theory and Object Kinetic Monte Carlo Models*

- 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.
- RT and OKMC models are similar kinetic models
  - can be used simulate the same phenomena
  - some of the details are handled quite 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

*in collaboration with C. Domain (EDF) and C. Becquart (U of Lille)

RT-OKMC: Inherent differences

Parameter or mechanism	Rate theory	Object kinetic Monte Carlo
solution method	deterministic	stochastic
time	explicit variable	inferred from processes and reaction rates
space	smeared, effective medium, possible multi-region RT	full spatial dependence
defect production	time and space-averaged, but c.f. Mansur's cascade diffusion model	discrete in time and space
sink strength, e.g. dislocations	explicit mathematical expression	inferred from fate of point defects
defect or sink density	essentially unlimited	limited (computationally) by simultion cell size, i.e. $N = 1/(x \cdot y \cdot z)$

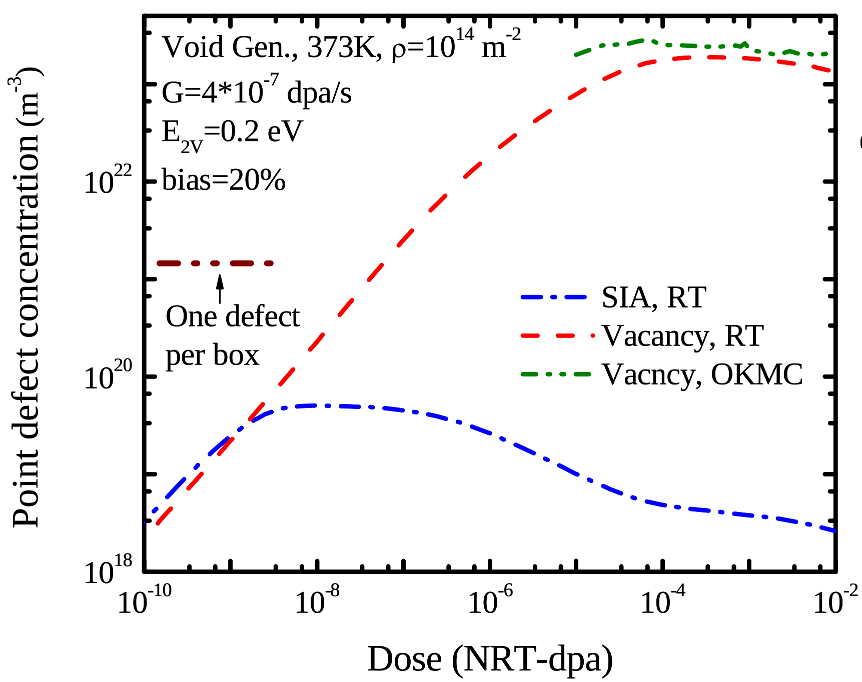
Implications for comparing simulation results: Making sure you are simulating the same problem

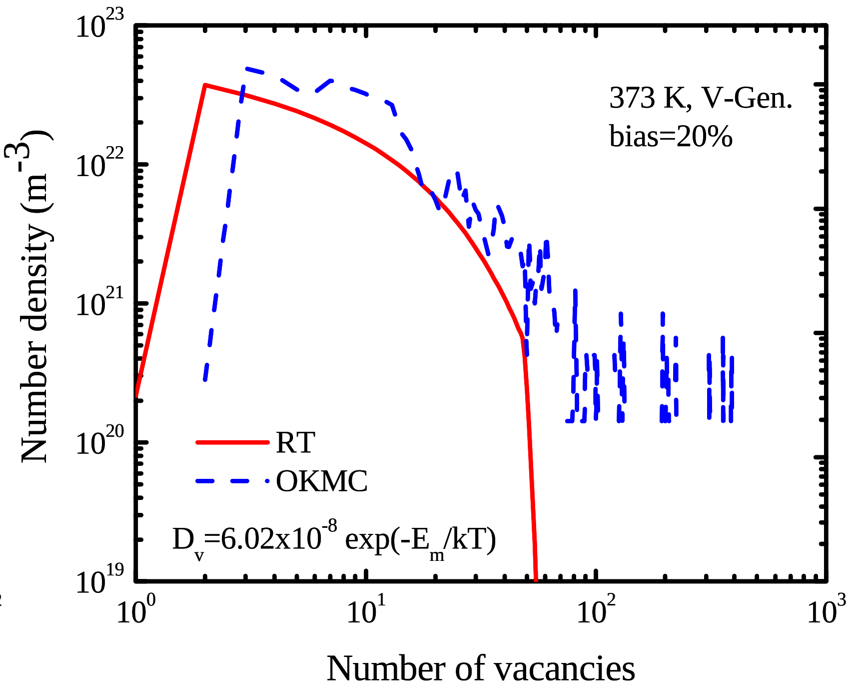
- deterministic vs. stochastic: not possible to do exact point-to-point comparisons
- spatial correlations when simulating cascade defect production: lost in RT, how handle in OKMC
  - electron irradiation with only FP production most straight forward
  - spatial relationship of point defects and clusters if use cascade results as input in OKMC
- minimum achievable defect/sink density in OKMC
  - e.g. for bcc iron, $x=y=z=300a_o$,

    $V=6.42\text{x}10^{-22}\ m^{-3}$, $1/V=1.57\text{x}10^{21}\ m^{3}$
  - limits a combination of maximum temperature and/or minimum displacement rate

Preliminary results

- bcc iron, G_{dpa}=1x10^{-6} dpa/s, up to 0.01 dpa, 100°C
- cascade production, 35% of vacancies in small (2,3) clusters, dislocation bias=20%
  - point defect concentrations and vacancy cluster size distribution, reasonable agreement with in-cascade vacancy cluster nucleation





Displacement rate (neutron flux level) effects in RPV steels

- issue of displacement rate effects on radiation-induced embrittlement of reactor pressure vessel steels remains unresolved: e.g.differences between recent alternate forms of RPV embrittlement correlation
- issue is relevant to:
  - difference between RPV surveillance positions and test reactor irradiations
  - BWR vs PWR vessels
  - damage attenuation through the RPV
- summary of model-based predictions of flux or displacement rate effects on radiation-induced hardening (yield strength changes)
- illustrate the range of possible/plausible effects in both low and high copper steels
- briefly describe model, key parameters, relevant mechanisms, and model predictions

RPV Embrittlement Model

A model using the kinetic rate theory was developed to describe the time-dependent evolution of point defects, point defect clusters, and copper precipitates (Stoller in ASTM STPs 1175, 1270, and 1325)

- Interstitial cluster formation can occur in the model by essentially classical nucleation (random collisions between single interstitials) or directly in the displacement cascade
- Vacancy clusters were treated as forming at a single size due to cascade collapse
- The results of molecular dynamics simulation studies were used to provide guidance for in-cascade clustering fractions and defect survival fractions.
- Copper precipitation model:
  - precipitate nucleation not modeled, N_{ppt}=f(Cu,T) and $r_{ppt}(0)$=0.25 nm
  - precipitate growth assuming diffusion limited kinetics

$$\frac{dr_{ppt}}{dt} = 4\pi\, r_{ppt}\, D^{j}_{Cu}\left(Cu^{o} - f_{Cu}\right)$$

- where: Cu^o is the initially available copper in the matrix and f_{Cu} is the amount of copper in CRP formed under irradiation

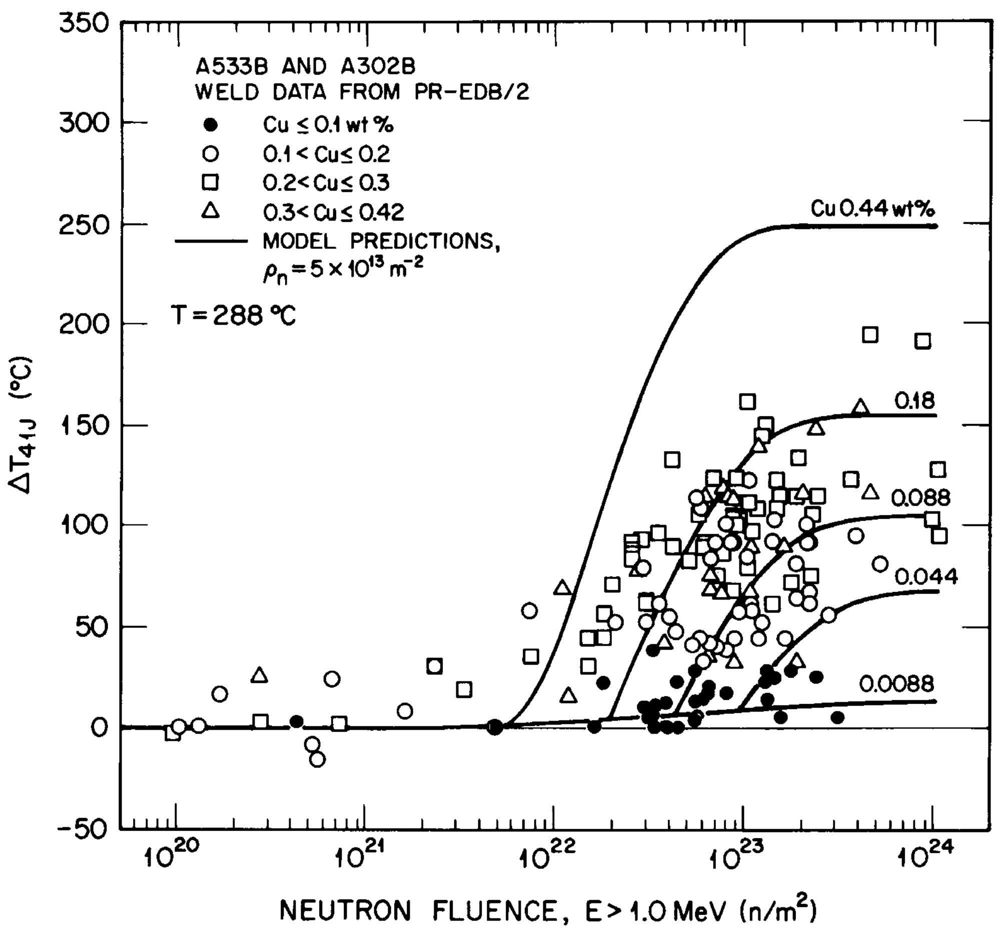
- D^j_{Cu} is the radiation-enhanced copper diffusion coefficient, = D_{Cu} (C_v/C_v^e)

- Strength change due to point defect clusters and copper precipitates
  - A simple dislocation barrier hardening model is used to calculate the shear stress increment required to cut through interstitial and vacancy type PDCs
  - Russell-Brown model (1972) used to compute hardening due to copper precipitates, based on modulus difference between precipitate and matrix
- Root-sum-square combination of individual contributions used to obtain total hardening:

$$\Delta\tau_{total} = \left(\Delta\tau^2_{icl} + \Delta\tau^2_{vcl} + \Delta\tau^2_{ppt}\right)^{0.5}$$

- Shear strength increments can be converted to corresponding changes in the uniaxial yield strength using the Taylor factor ($\Delta\sigma_y$=3.06*$\Delta\tau$), and yield strength changes can be related to Charpy shifts using correlations from the literature (e.g. Odette, et al; Williams, et al.) ΔT_{41}~[0.5-0.65]*$\Delta\sigma_y$

TYPICAL MODEL PREDICTIONS



reasonably good agreement with surveillance data from PR-EDB,
- e.g. copper and fluence dependence
- considerable data scatter

stronger copper dependence in model since all copper treated as available for precipitation

rather abrupt transition, too simple copper precipitate model, i.e. no nucleation component

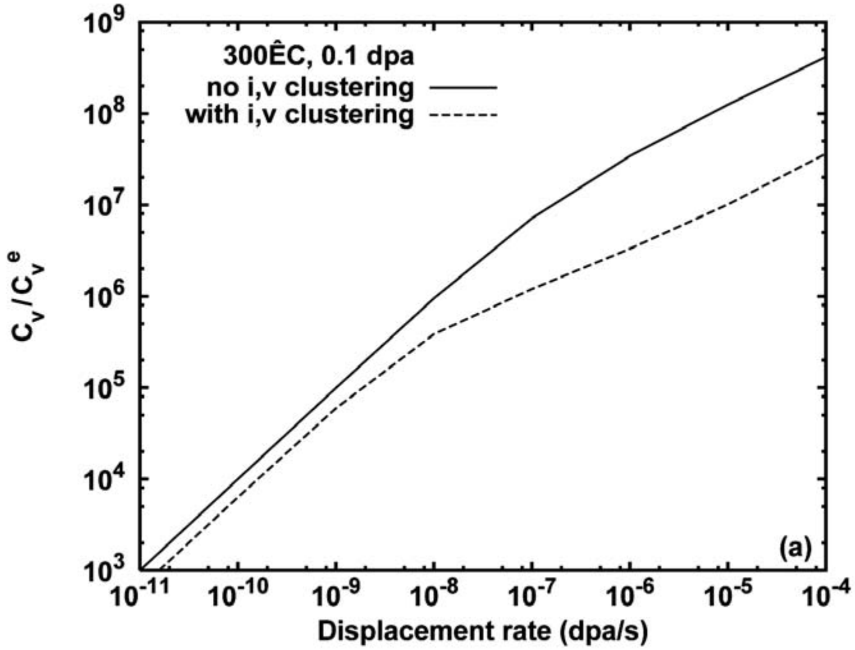
- “typical” material and model and parameters

BASIC MECHANISMS RESPONSIBLE FOR A DISPLACEMENT RATE EFFECT

Radiation-induced property changes are driven by the excess point defect fluxes created by displacive irradiation.

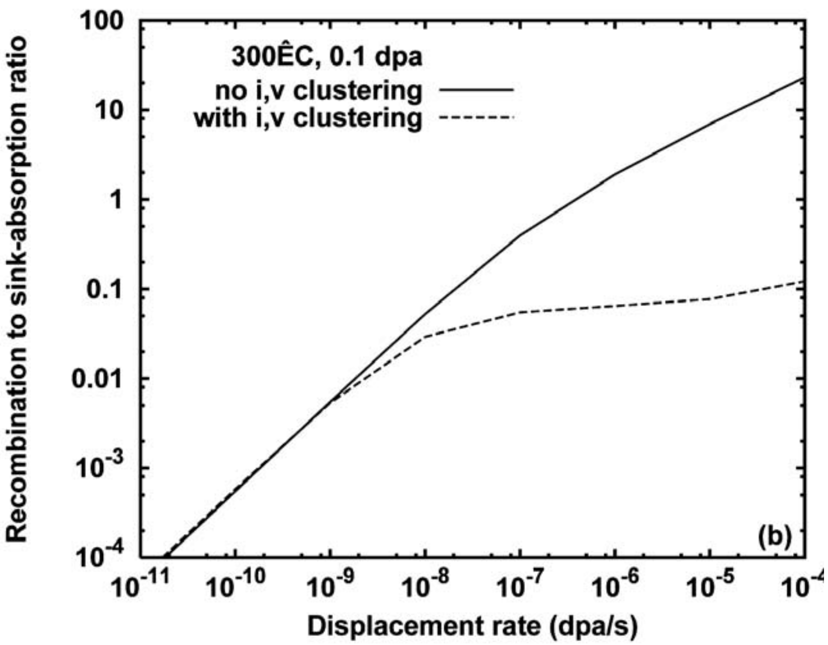
- Details of microstructural evolution and effects such as solute segregation are determined by the transport and fate of mobile point defects and solutes
- Displacement rate effects in RPV steels arise primarily from two processes
  - the competition between formation and dissolution of unstable defects (this component is nearly inseparable from the effects of irradiation temperature)
  - the influence of the displacement rate on radiation-enhanced diffusion
- Hardening from point defect clusters is most strongly influenced by the first process, and from copper precipitates by the second
- However, when the unstable defects provide a significant sink for mobile point defects, they will have an impact on radiation-enhanced diffusion
- Thus, an increase in displacement rate may lead to either an increase or decrease in hardening

Displacement rate effect: vacancy supersaturation and matrix recombination

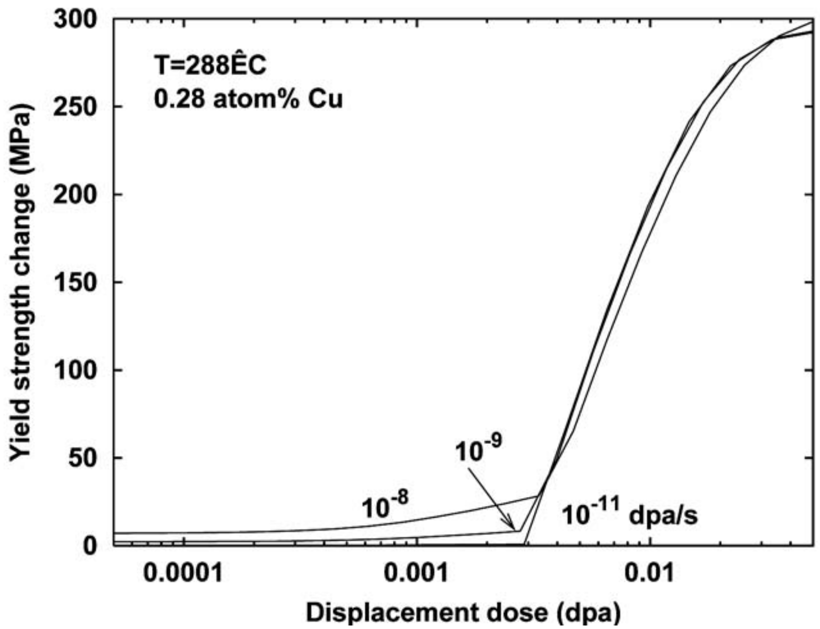


- rate effect reduced by high sink density with in-cascade clustering

- alternate curves show effect of including in-cascade point defect clustering



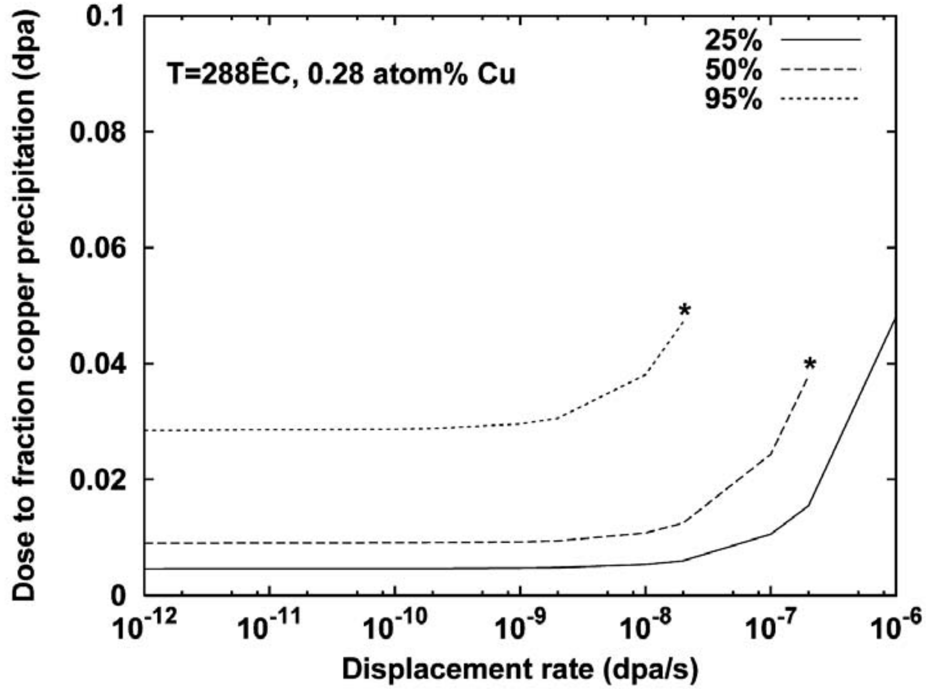
MODEL PREDICTIONS: fluence dependence of hardening)



Note: for typical displacement cross section of 1500 barn, $1x10^{19}$ n/cm^2 = 0.015 dpa

- In high-copper steels, primary effect of lower displacement rate is a reduced fluence to initiate hardening from copper-rich precipitates. Little effect on peak hardening.
- At low fluences, hardening is reduced at lower displacement rates. Can lead to a crossover at intermediate fluences

MODEL PREDICTIONS: dose required to reach a given level of copper precipitation

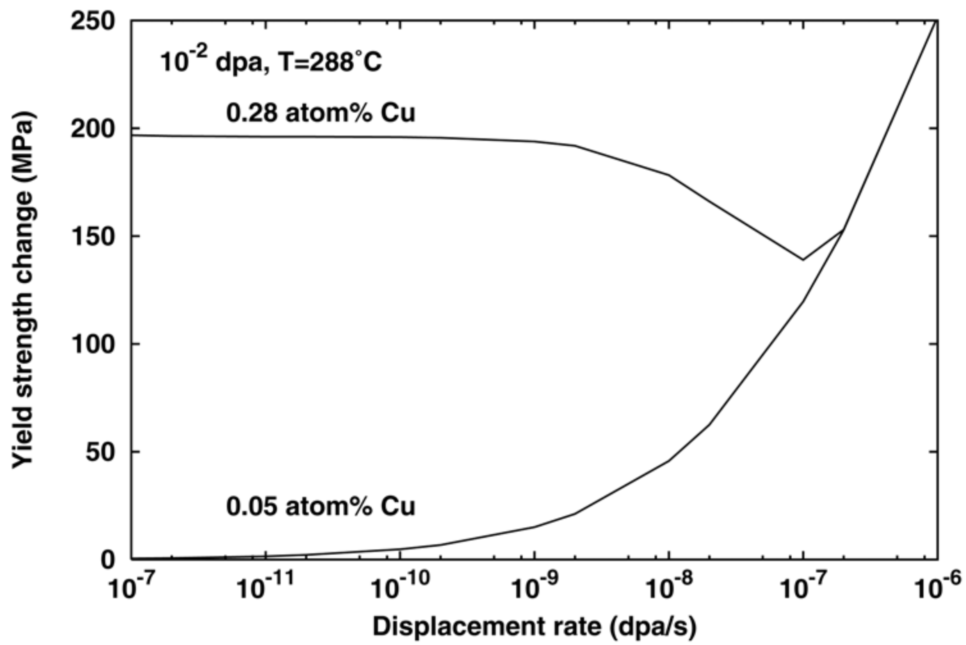


* 50 and 95% precipitation not reached by 0.05 dpa

- threshold dose reduced at lower displacement rates
- dose-rate independent region at lowest displacement rate
- transition to dose-rate independence is moved to lower displacement rates for higher fractional precipitation

MODEL PREDICTIONS:

Predicted yield strength change at 10^{-2} dpa for low and high copper steel



- In low-copper steels, hardening is reduced at lower displacement rates
- Effect of displacement rate stronger than observed in most low-copper data

Summary of Rate Effects in RPV Embrittlement

Relatively simple kinetic model illustrates the effect of variations in flux on radiation-induced hardening

- the predicted influence of damage rate on radiation-induced hardening depends on several other material and irradiation variables:
  - copper content
  - neutron fluence
  - flux range
  - temperature and materials parameters not discussed here, e.g. surface energy, assumptions in hardening models
- depending on which values apply, a change in flux may lead to either an increase or a decrease in hardening
- the absolute magnitude of the flux effect, and the flux range with the greatest predicted sensitivity depends on the details of the model and the chosen parameters
- model predictions are consistent with a modest effect of neutron flux for the range of fluxes of interest to LWR RPV at ~290°C

Summary: Status and opportunities in radiation effects modeling and simulation

Substantial progress in understanding and predicting the response of materials to displacive irradiation has been achieved through:

(1) development of large experimental databases and

(2) recent advances in theory and computational modeling

- progress enhanced by close coupling of theory/modeling/simulation and experimental programs
- increasing scale of *ab initio* calculations and large scale atomistic simulations identify new mechanisms and provide underlying support to multiscale modeling framework
- more detailed mesoscale models enabled, improved parameter definition
- “real” multiscale development and linking still needed where it makes sense

Applicable to:

- advanced fission, Gen-IV and GNEP, fission reactor materials
- fusion reactor materials
- other applications for advanced materials