U.S. DEPARTMENT OF ENERGY

NEAMS
NUCLEAR ENERGY ADVANCED MODELING & SIMULATION PROGRAM

Meso-Scale Phase-Field Model and Bridge to Macroscopic Property Predictions

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Computational Sciences and Mathematics Division

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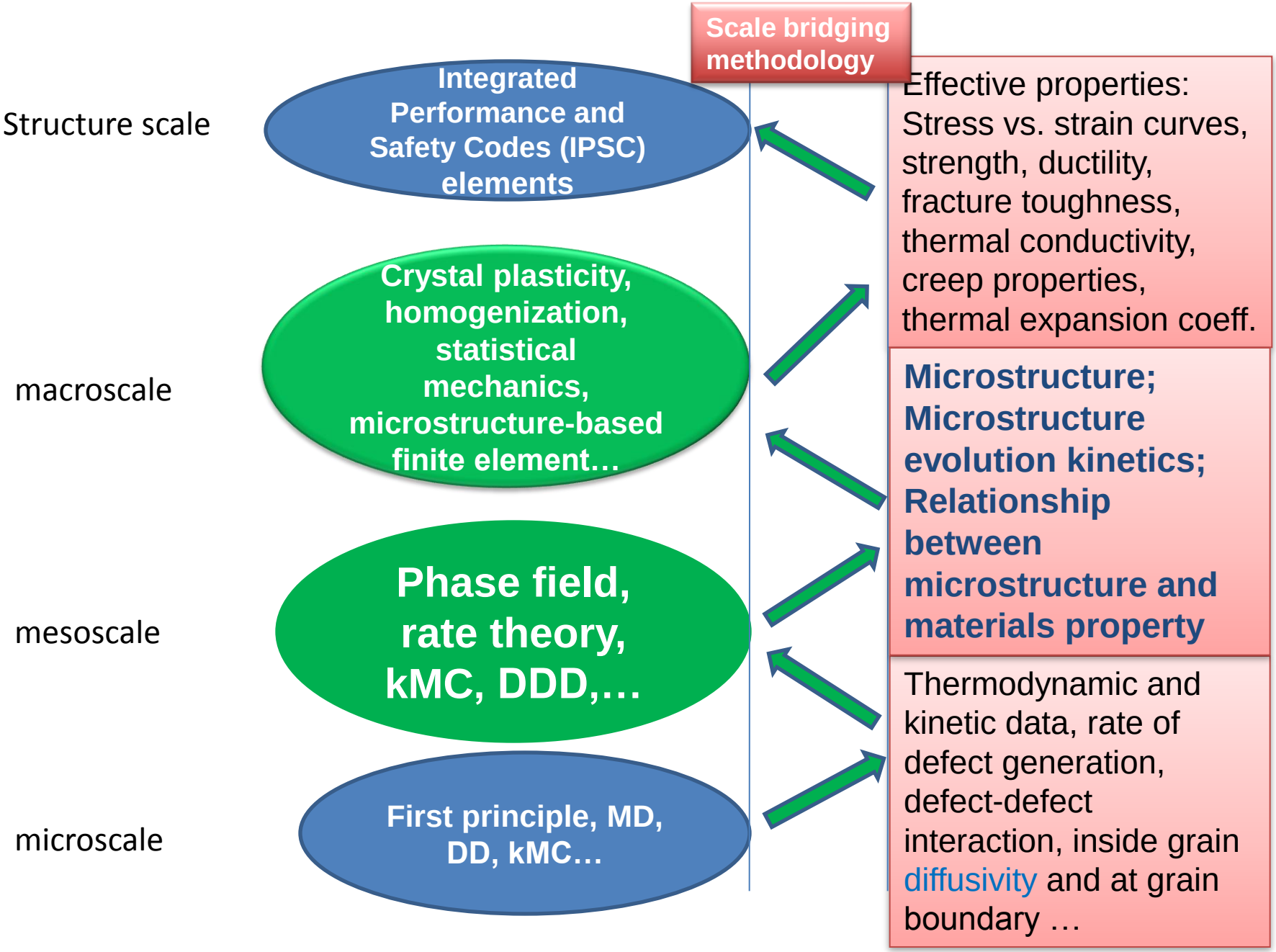
FMM Objectives

- Provide cross cutting fundamental models and methods to various IPSCs
- Develop fundamental materials modeling capability starting from the molecular level
- Provide upscaling methods
- Provide new methods and algorithms
- Provide single point of contacts for IPSCs
- Collect requirements from IPSCs
- Provide interactions with NEUP projects
- Provide benchmark and comparison of various modeling techniques at various scales for various phenomena

FMM Technical Approach



FY10 Accomplishments

- Developed meso-scale phase-field modeling framework to simulate material degradation under irradiation:
  - Void migration
  - Volume swelling
- Built and evaluated various scale bridging methodologies:
  - Meso-scale microstructure representation and reconstruction
  - Upscaling of thermal properties from meso-scale to macro-scale:
    - Waste form - Glass with different waste loading
    - Fuel - Isotropic and anisotropic UO2 with evolving gas bubbles at grain boundaries
- Published/submitted one level 4 NEAMS milestone report and eight journal articles

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Phase-field Approach

Total Free Energy

Total free energy may include bulk chemical free energy, interfacial energy, elastic energy, magnetic energy, and electrostatic energy under applied external fields such as temperature, applied stresses, electrical and/or magnetic fields. Thermodynamic and kinetic data can be obtained by atomistic simulations and experiments.

$$E = \int_V [\, f(c_1, c_2, \cdots, c_N, \eta_1, \eta_2, \cdots, \eta_P, T) dV$$

Bulk chemical free energy

$$+ \int_V \sum_{n=1}^{N} \alpha_n (\nabla c_n)^2 + \sum_{p=1}^{P} \beta_{ij} \frac{\partial \eta_p}{\partial r_i} \frac{\partial \eta_p}{\partial r_j}] dV$$

Interfacial energy

$$+ \int_V \int_{V'} G(\boldsymbol{r} - \boldsymbol{r}') dV dV'$$

Long-range interaction

Conserved variables:

$$c_1, c_2, \ldots, c_N$$

Non-conserved variables:

$$\eta_1, \eta_2, \ldots, \eta_P$$

Gradient energy coefficients:

$$\alpha_n, \beta_{ij}$$

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Phase-field Approach (cont.)

Equations Governing Microstructure Evolution

Cahn-Hilliard equations for conserved fields (concentrations)

Defect generation, defect reaction, nucleation,…

Ginzburg-Landau (or Allen-Cahn) equations for non-conserved fields (order parameters)

Defect generation, defect reaction, nucleation,…

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together with

- ❑ **Chemical equilibrium equations**
- ❑ **Mechanical equilibrium equations**
- ❑ **Electric field**
- ❑ **Boundary conditions**

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Void Migration under Temperature Gradient

- Two phases coexist: void and matrix
- Two kinds of defects: vacancies and interstitials
- Generation of irradiation defects
- Recombination between vacancies and interstitials
- Resolution of vacancies from voids
- Temperature field

Two variables $c_{Vac}(\mathbf{r},t)$ and $c_{Int}(\mathbf{r},t)$ are used to describe the distribution of vacancies and interstitials. Their evolutions are described by the following Cahn-Hilliard equations (dimensionless):

$$\frac{\partial c_{Vac}}{\partial t} = M_{Vac}\nabla^2\left(\frac{\partial F}{\partial c_{Vac}} - \kappa_{Vac}\nabla^2 c_{Vac}\right) + \dot{g}_{Vac} - \dot{\gamma}_0 c_{Vac} c_{Int}$$

$$\frac{\partial c_{Int}}{\partial t} = M_{Int}\nabla^2\left(\frac{\partial F}{\partial c_{Int}} - \kappa_{Int}\nabla^2 c_{Int}\right) + \dot{g}_{Int} - \dot{\gamma}_0 c_{Vac} c_{Int}$$

Li, et al. *J. Nuclear Mat.*, accepted. 2010.

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Chemical Free Energy of a System with Defects

$$F(c_{Vac}, c_{Int}, T) = F_1(c_{Vac}, T) + F_2(c_{Int}, T)$$

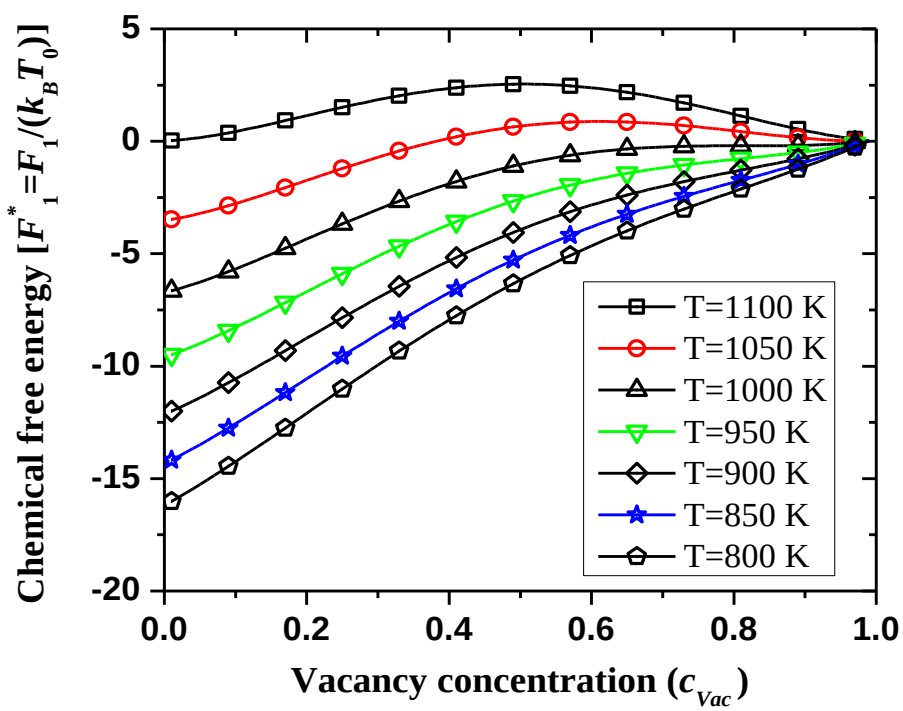
$$F_1(c_{Vac}, T) = k_B T\left[(1-c_{Vac})\ln(1-c_{Vac}) + c_{Vac}\ln c_{Vac}\right] + f_1(c_{Vac}, T)$$

$$F_2(c_{Int}, T) = k_B T\left[(1-c_{Int})\ln(1-c_{Int}) + c_{Int}\ln c_{Int}\right] + a c_{Int}$$

$$f_1(c_{Vac}, T) = b_4 c_{Vac}^4 + b_3 c_{Vac}^3 + b_2 c_{Vac}^2 + b_1 c_{Vac} + b_0$$

For a given material, the chemical free energy can be fitted by considering the following thermodynamic properties:

(1) equilibrium vacancy concentration c_{Vac_M} in the matrix phase
(2) equilibrium vacancy concentration c_{Vac_V} for the void phase
(3) F_1 has common tangent at c_{Vac_M} and c_{Vac_V}
(4) spinodal point c_{Vac_s}
(5) $F(c_{Vac_M}, T)$ is temperature dependent
(6) equilibrium interstitial concentration c_{Int} is very small, 0.02%
(7) a is interstitial formation energy



These parameters link to lower scale simulations, e.g., MD

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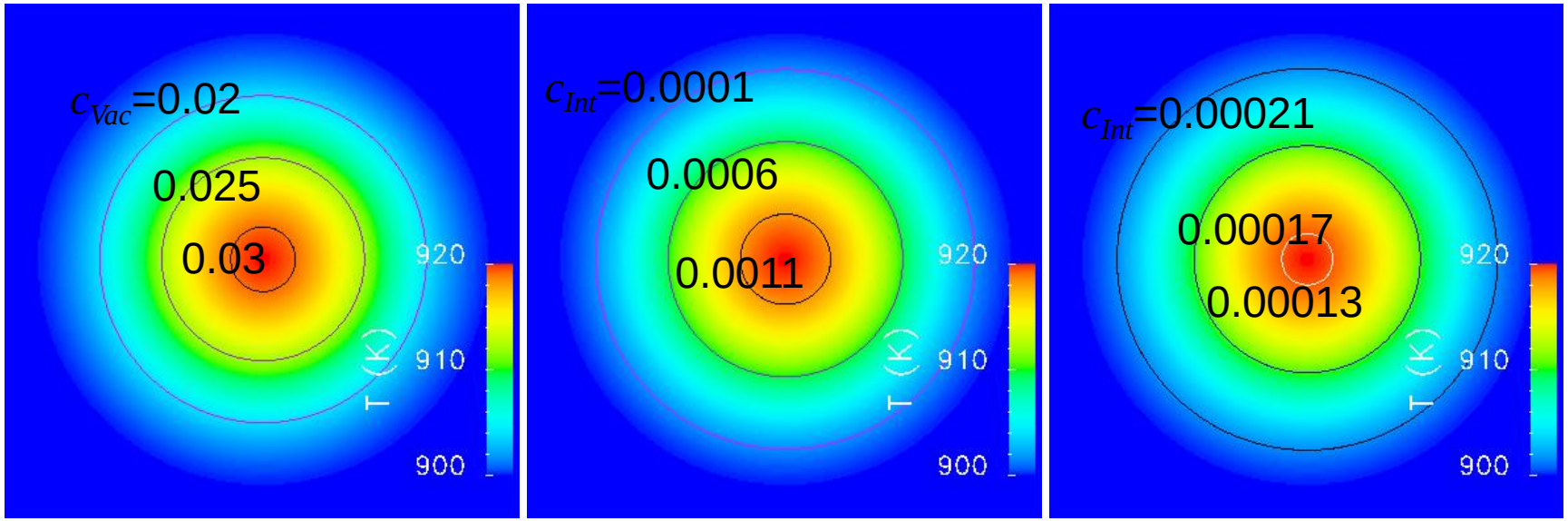
Soret Effect: Vacancy and Interstitial Migration under Temperature Gradient



- **Vacancies migrate up to higher temperature**
- **Interstitials migrate up to higher temperature when interstitial formation energy is constant, i.e., $a=a_0$**
- **Interstitials migrate down to lower temperature when interstitial formation energy increases with temperature as $a=a_0+a_1k_BT$**

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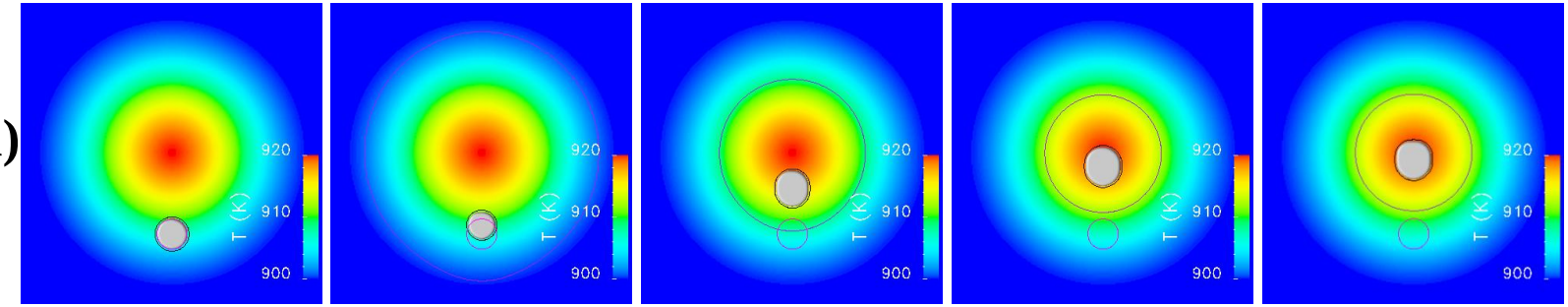
Single Void Migration in a Temperature Field

(a)



t^*=0 t^*=20 t^*=100 t^*=200 t^*=464

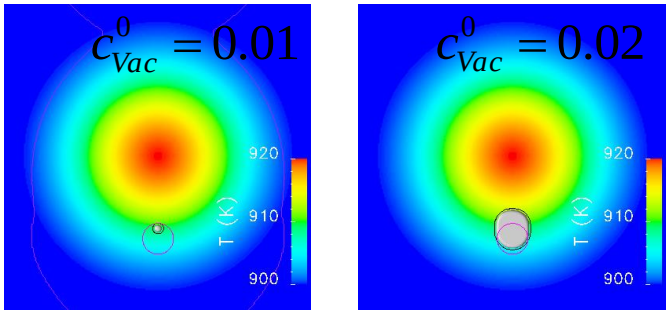
r_0*=12

$c^0_{Vac} = 0.015$

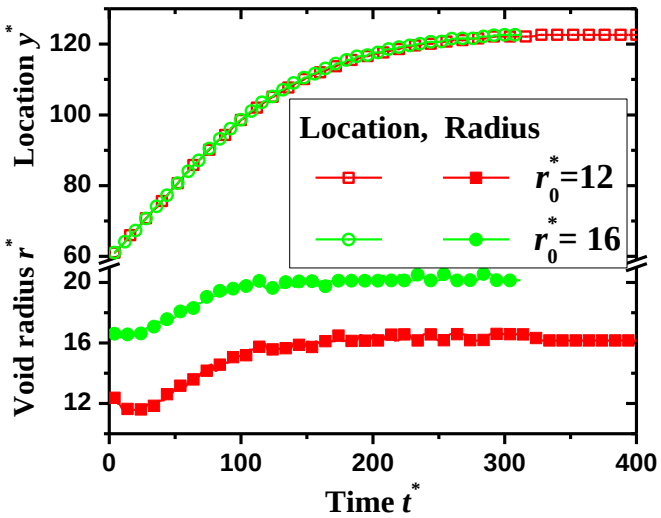
(b)

r_0*=12

t*=20

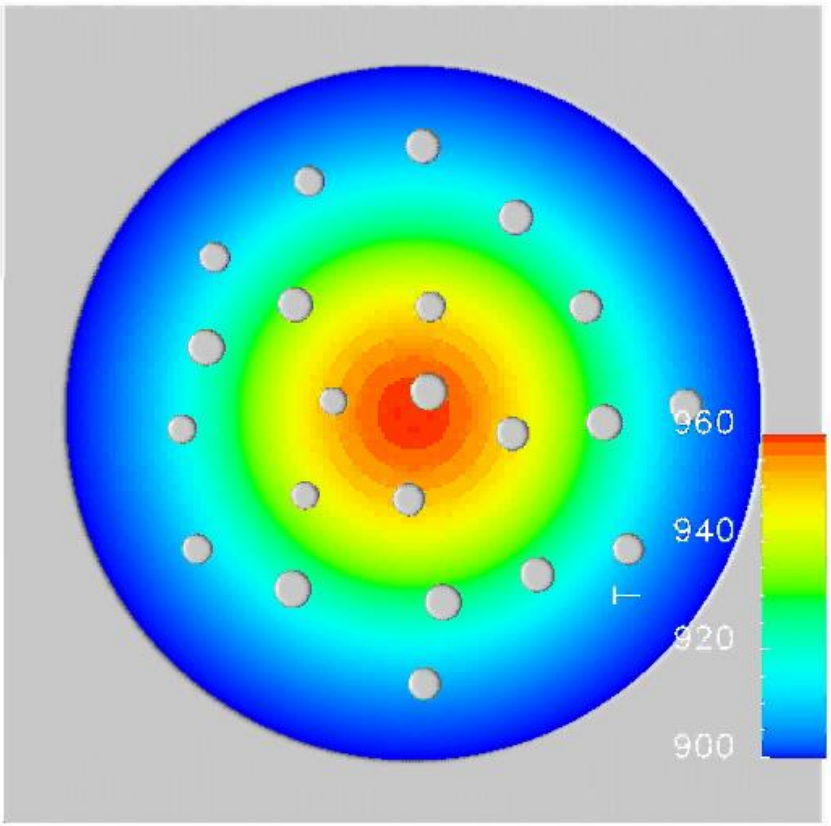


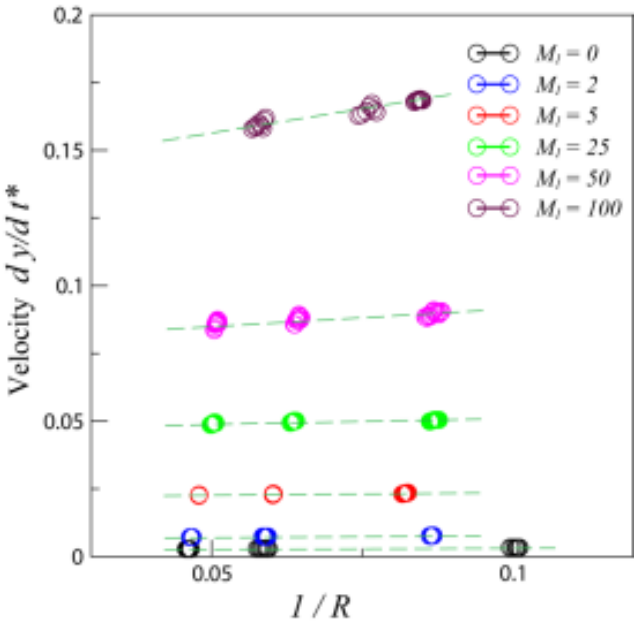
(c)



- Void migrates up to higher temperature
- Void cannot be stabilized when the vacancy concentration in the matrix is too low
- Larger matrix vacancy concentration speed up void growth
- Void migration mobility is independent of its size and vacancy concentration (bulk diffusion)

Central Hole Formation and Surface Diffusion





$$M^*(c) = M_0^*\left(1 + M_1\left[\exp\left(c_{Vac}\right) - 1\right]\right)$$

- **Voids migrate from low T to high T region**
- **Predicted size dependence of void migration velocity**

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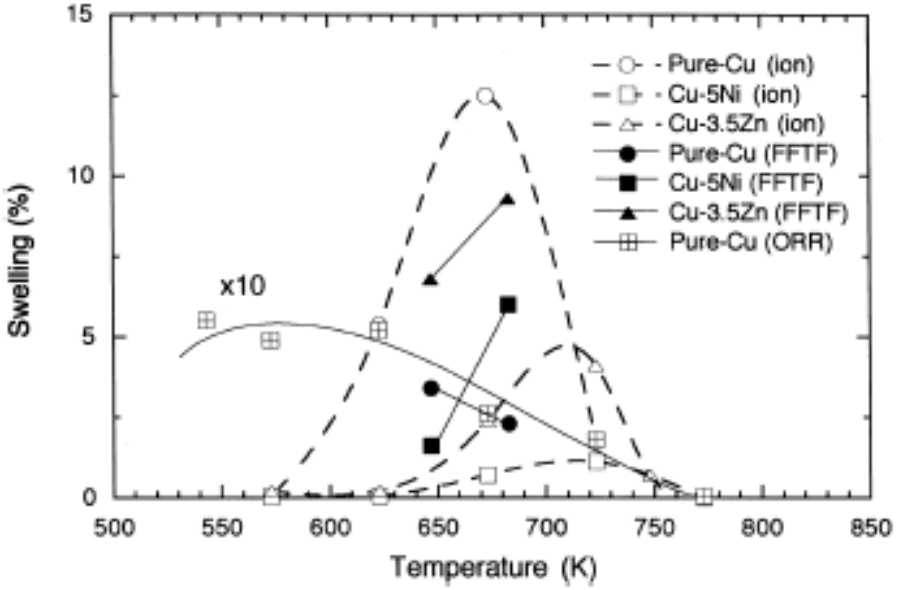
Volume Swelling in Irradiated Materials



T. Muroga, JNM, 258(1998)130

- Temperature
- Void nucleation
- Defect generation
- Defect mobility
- Defect reaction
- Stresses
- Resolution
- Solute, dislocation, precipitate, grain boundary

All affect the swelling kinetics

S. I. Porollo, JNM, 378(2008)17

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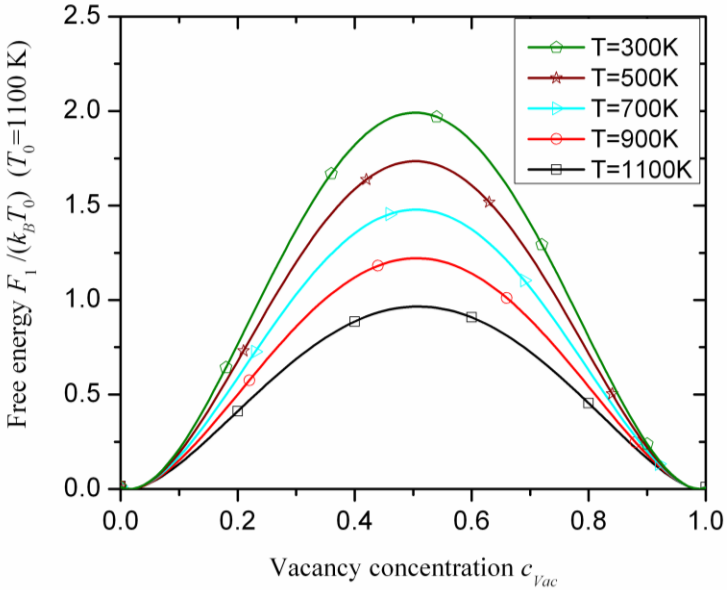
Free energy, Interfacial Energy & Mobility Used in Phase-field Simulations

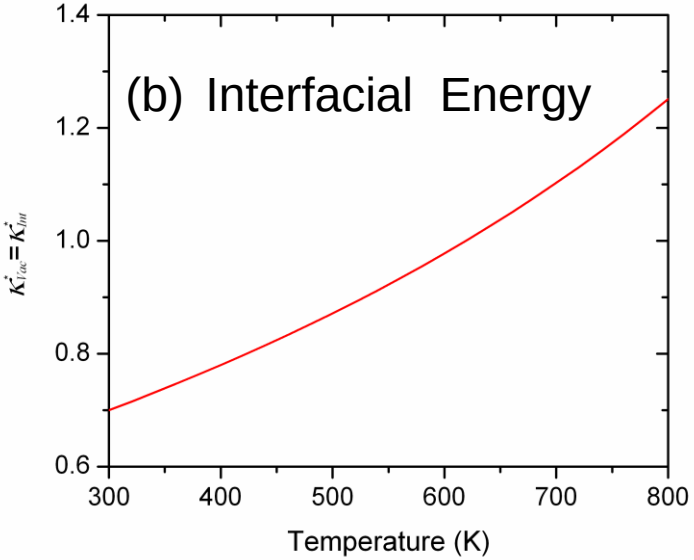
(a) Free Energy

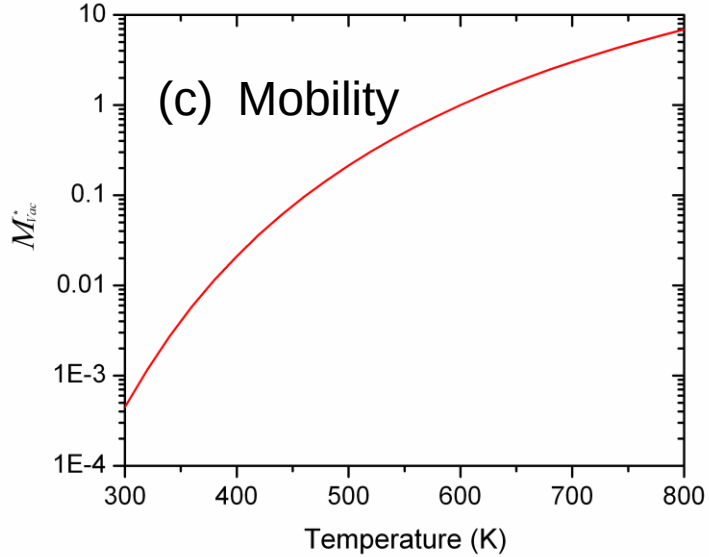
$$F(c_{Vac}, c_{Int}, T) = F_1(c_{Vac}, T) + F_2(c_{Int}, T)$$

$$F_1(c_{Vac}, T) = k_B T\left[(1 - c_{Vac})\ln(1 - c_{Vac}) + c_{Vac}\ln c_{Vac}\right] + b_4 c_{Vac}^4 + b_3 c_{Vac}^3 + b_2 c_{Vac}^2 + b_1 c_{Vac} + b_0 \ ,$$

$$F_2(c_{Int}, T) = k_B T\left[(1 - c_{Int})\ln(1 - c_{Int}) + c_{Int}\ln c_{Int}\right] + a c_{Int}$$

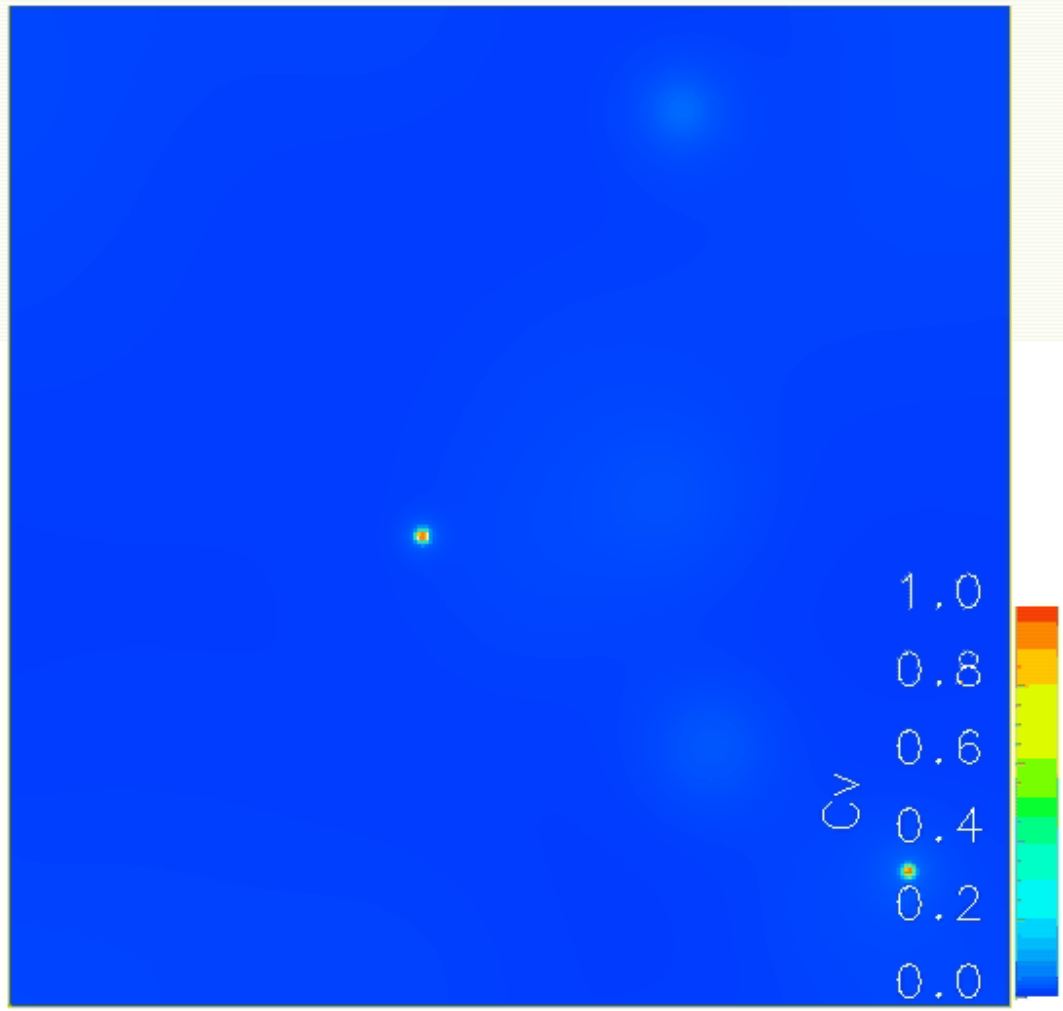






Li, et al. *J. Nuclear Mat.*, submitted. 2010.

Evolution of Void Structure at T=640K



Simulation conditions:

Temperature: 640K

Initial vac. concentration: 0.01

Initial int. concentration: 0.005

Generation rate: 0.004/ t_0

Recombination coefficient: 4.8/ t_0

Vacancy mobility: 1.62

Interstitial mobility: 13.5

Vacancy cluster generation rate: 9.6 per t_0

(dimensionless qualities)

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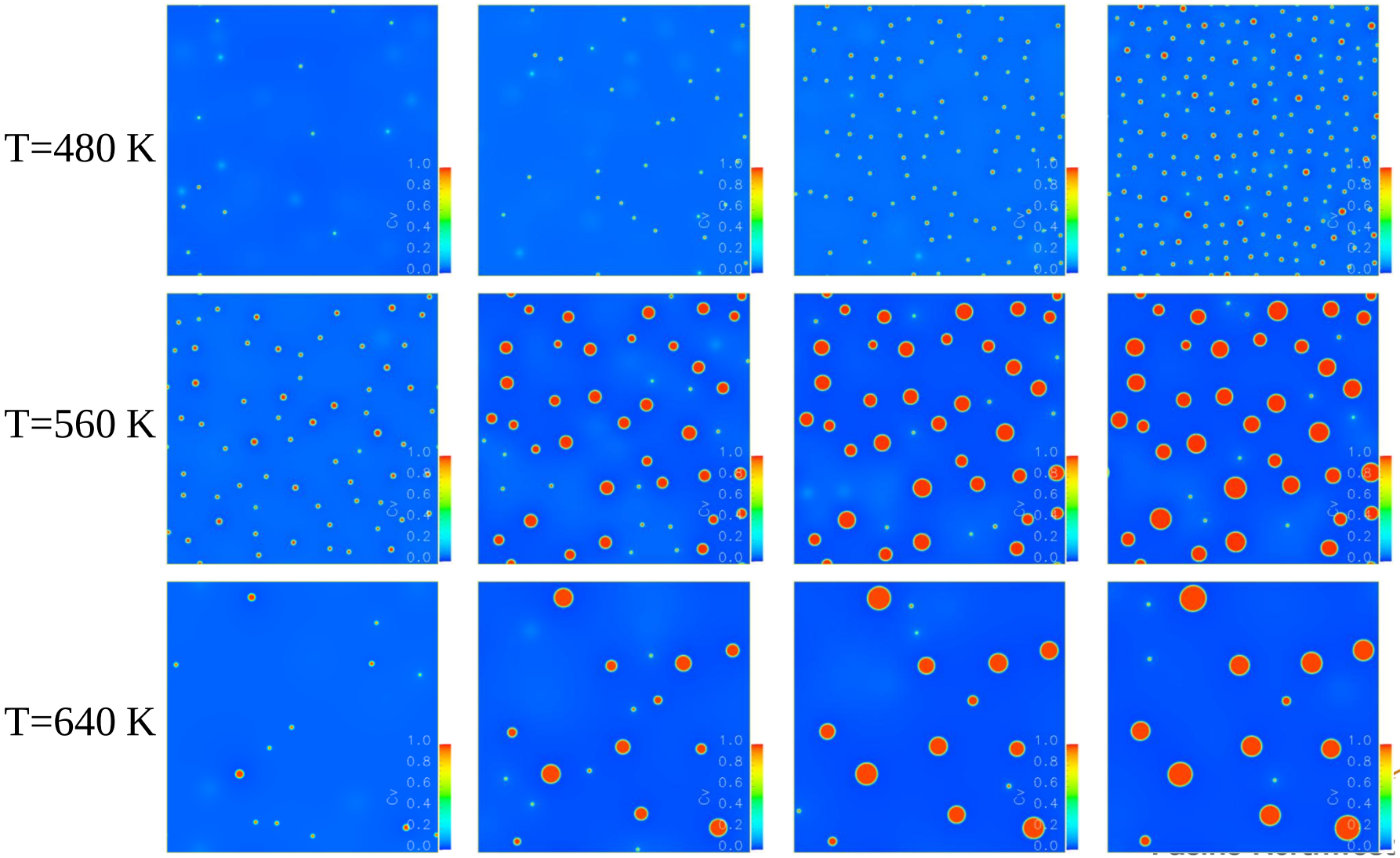
Snapshots of Void Structure Evolution at Different Temperatures



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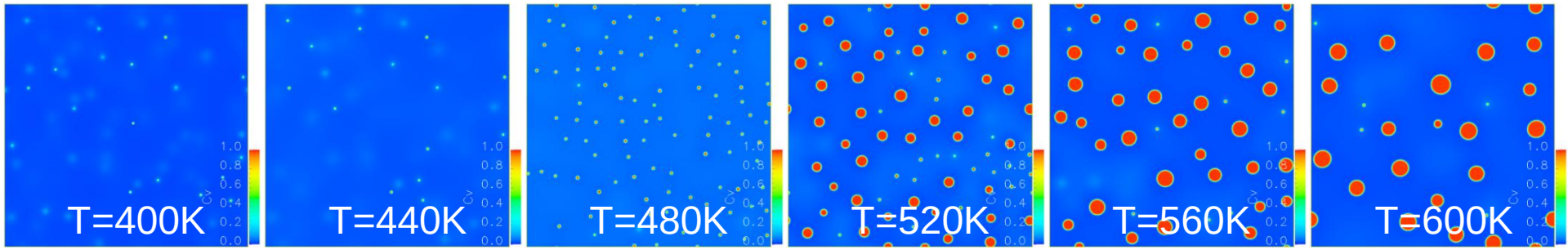
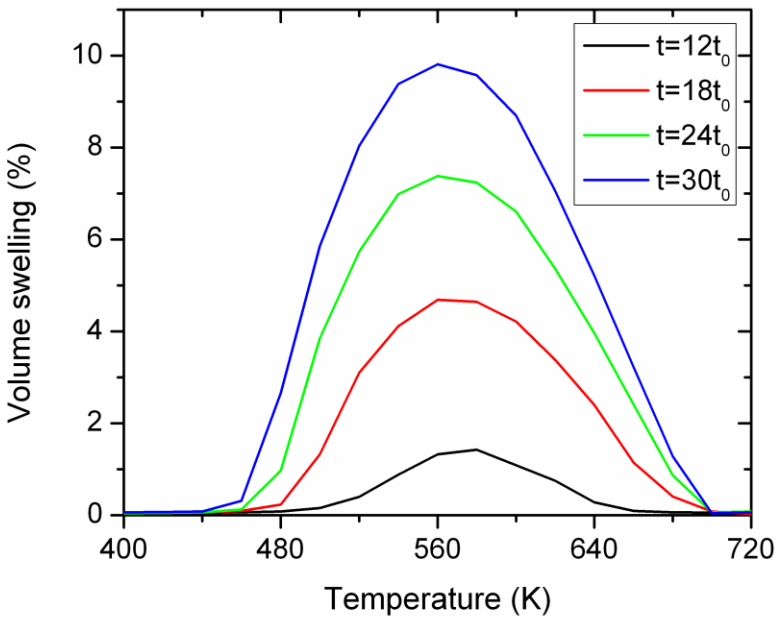
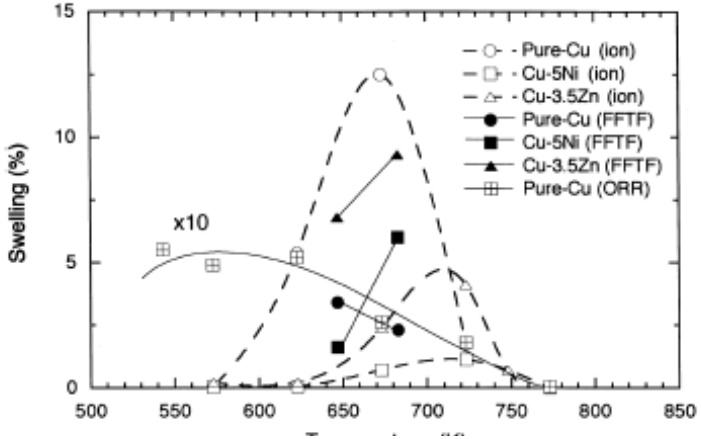
Predicted Void Microstructures and Void Volume Fraction Evolution

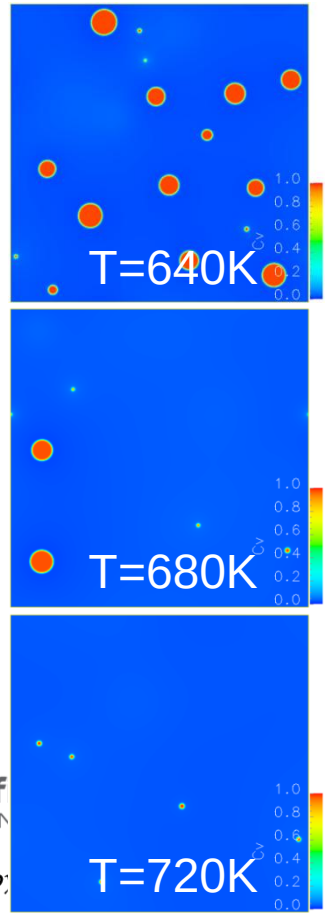


At time t=24t$_0$





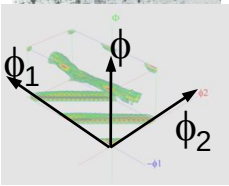
T. Muroga, JNM 258 (1998) 130



Microstructure Representation and Reconstruction

Multiple Modality
Multiple resolution

Spatial structural information

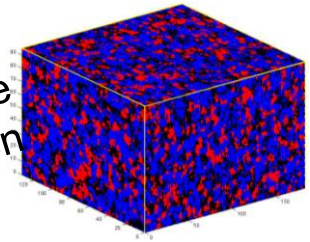


Statistical structural information

data reduction
data fusion

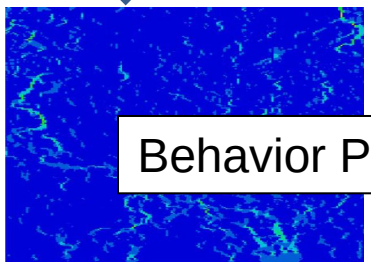
Structure descriptors

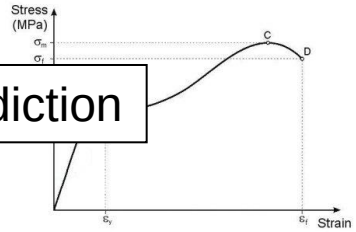
Signature microstructure reconstruction



Direct Upscaling

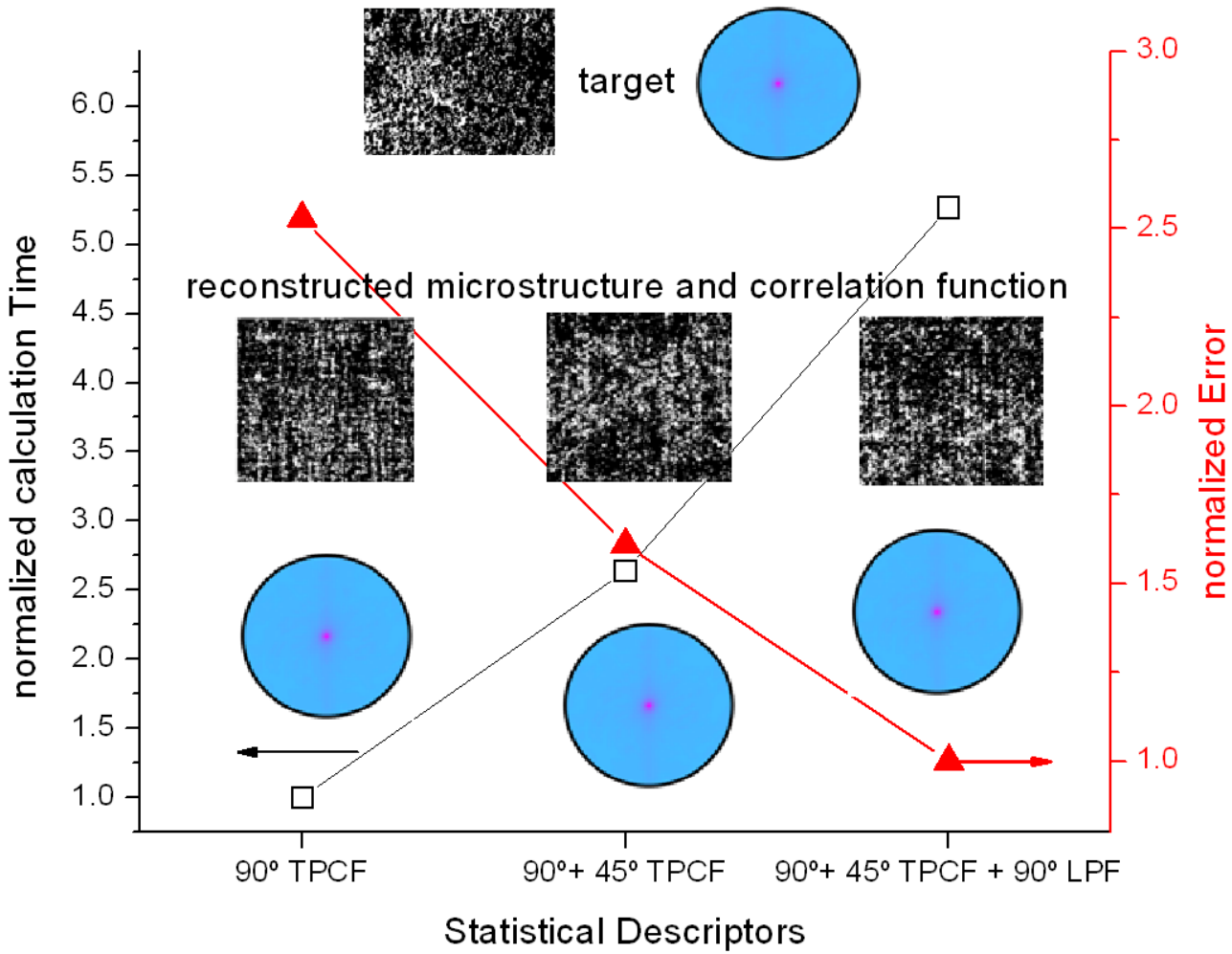
Indirect Upscaling





Behavior Prediction

Accuracy and Efficiency in Microstructure Reconstruction



Tschopp, et al. *Comparison of reconstructed spatial structures using different statistical descriptors. Computational Materials Science*. submitted.

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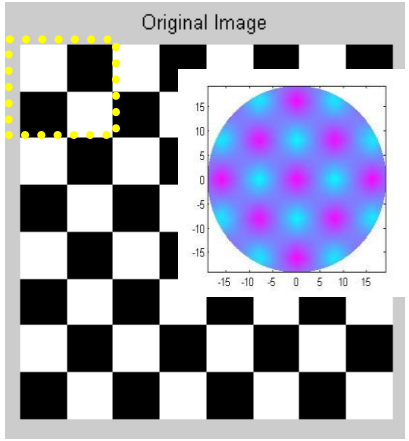
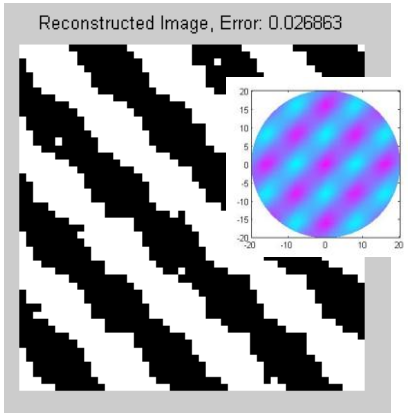
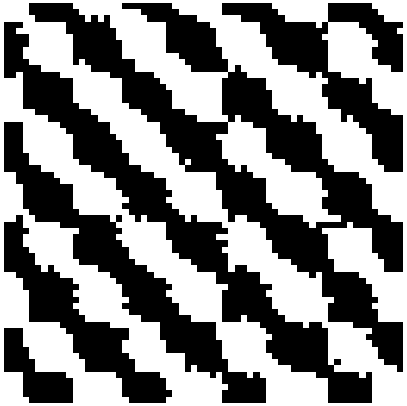
Microstructure Reconstruction -- Accuracy and Efficiency



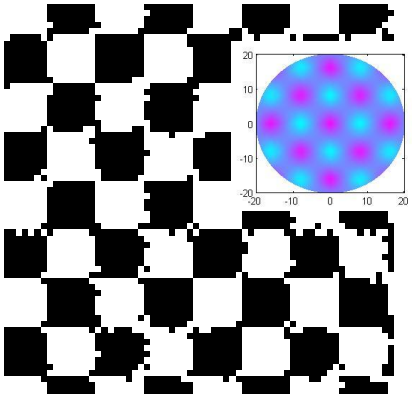
Target



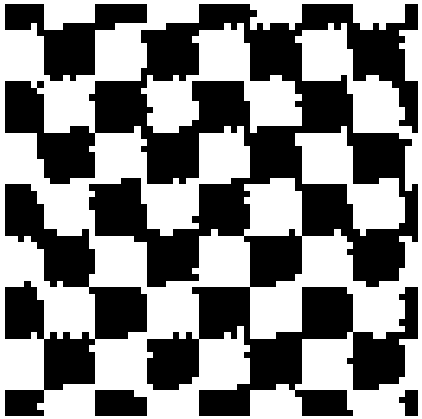
S2 on x and y directions



S2 on x and y directions
+L2 on x and y directions

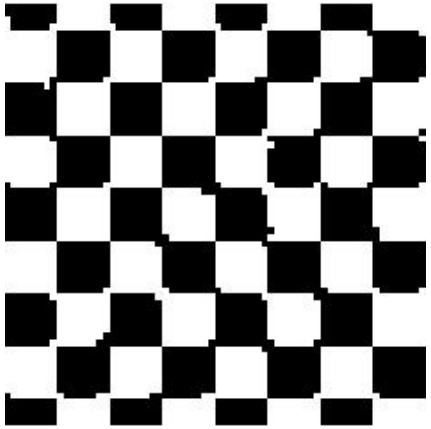


S2 on x and y directions
+2 diagonal directions



S2 on x and y directions
+2 diagonal directions
+L2 on x and y directions

error=0.01



S2 on x and y directions
+2 diagonal directions
+L2 on x and y directions

error=0.001

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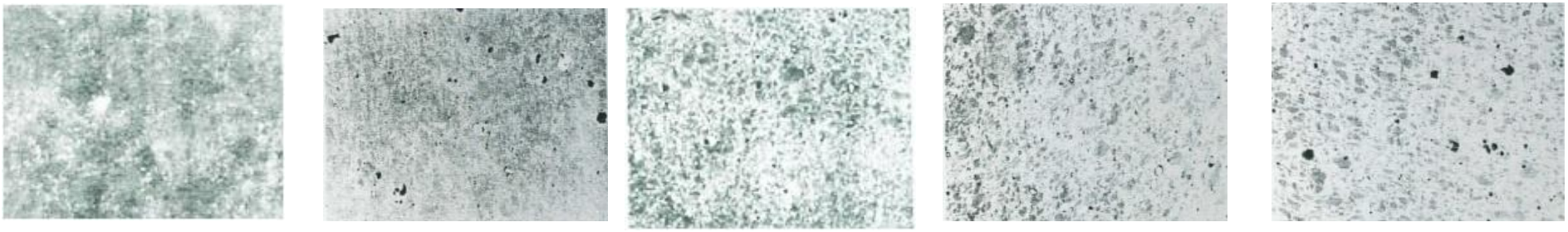
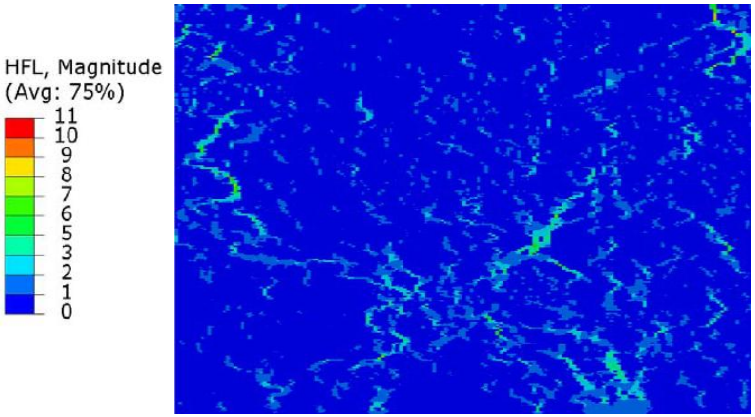
Assessment of Upscaling Methods

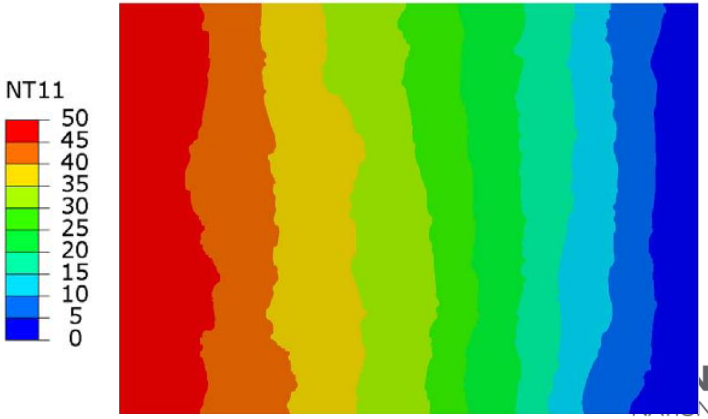
SG7-HLW glass loaded with nuclear waste



5% 10% 15% 25% 35%

S.Gahlert and G. Ondracek, in Radioactive Waste Forms for the Future, eds. W. Lutze and R.C. Ewing (NorthHolland, Amsterdam, 1988)





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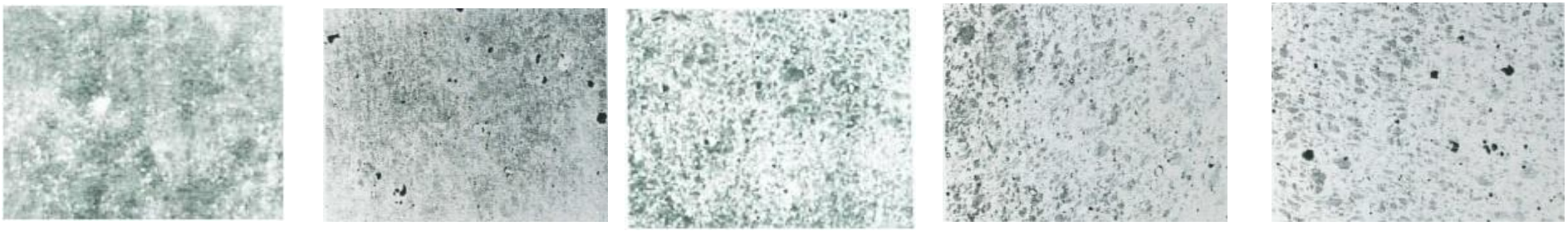
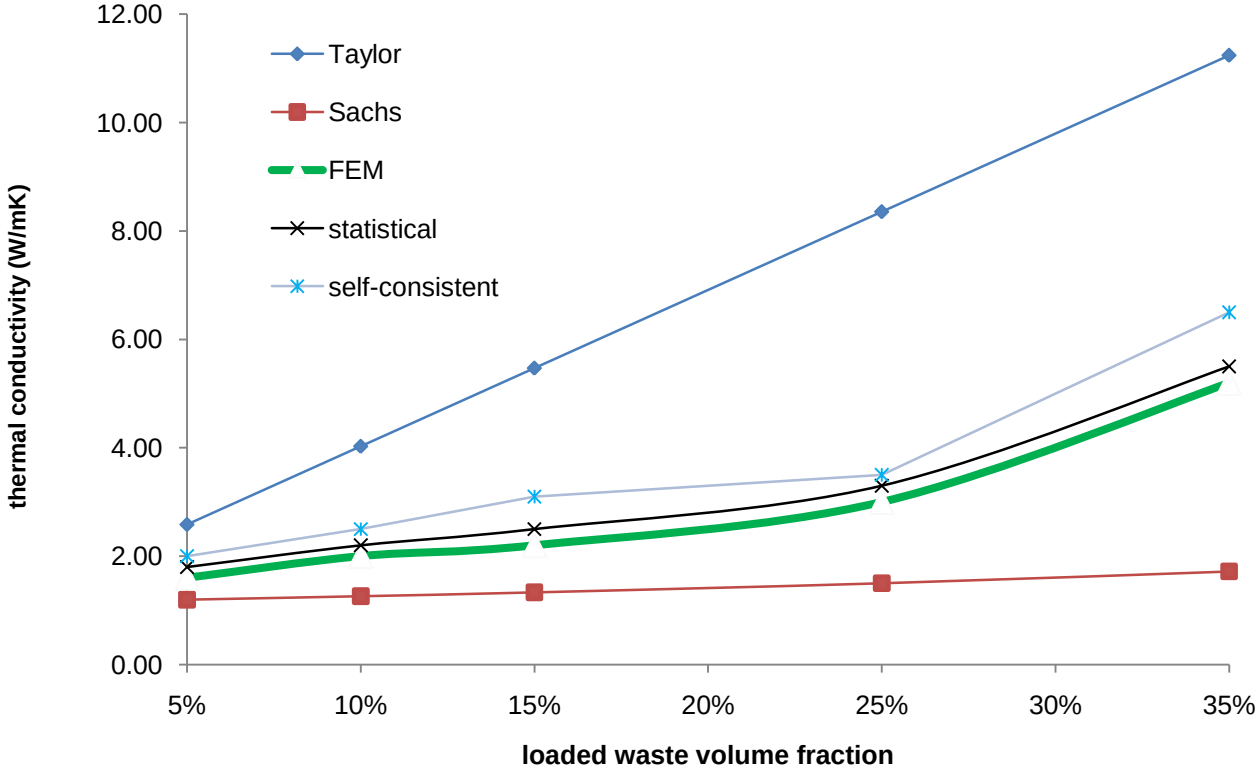
Example 1 - Assessment of Upscaling Methods for Waste Glass



5% 10% 15% 25% 35%



accuracy

FEM

Statistical

Self consistent

Bound method

efficiency

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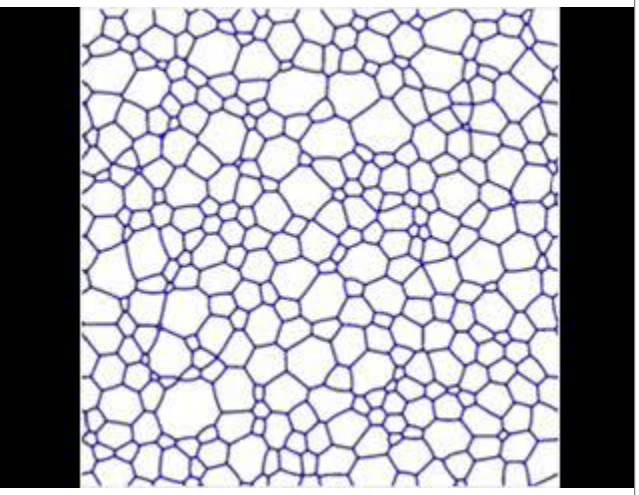
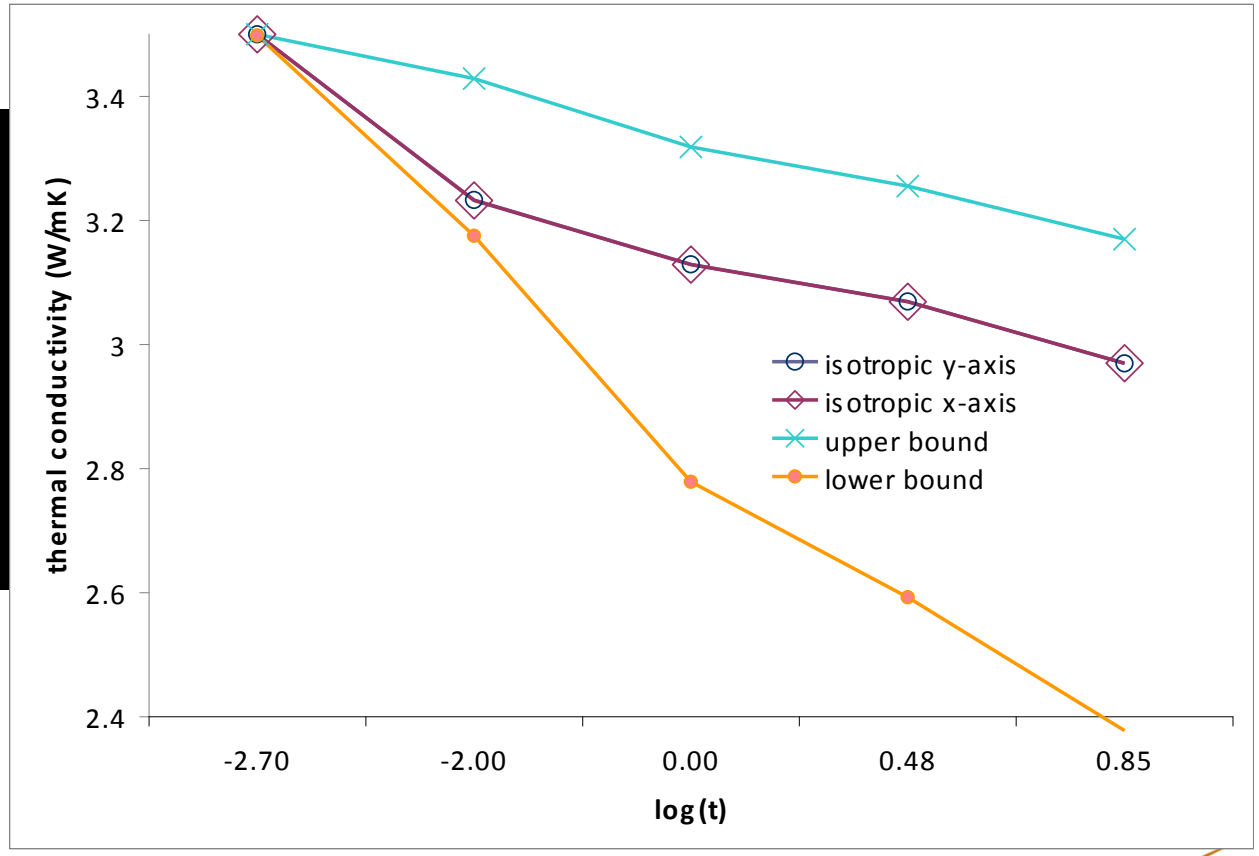
Example 2: Assessment of Upscaling Methodologies - From Phase field Model to Continuum Properties

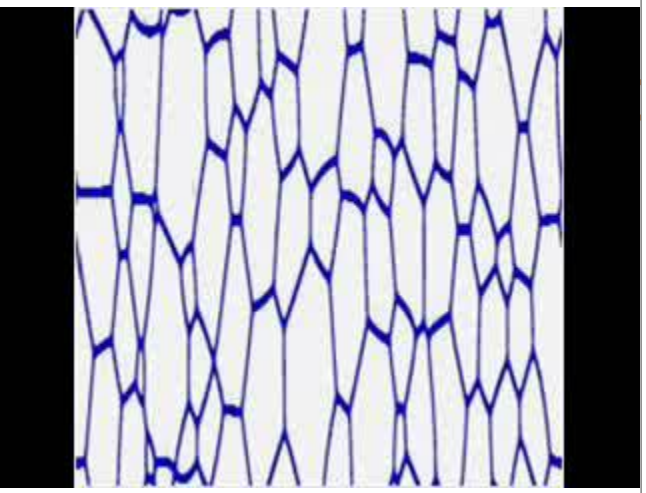


UO2 example

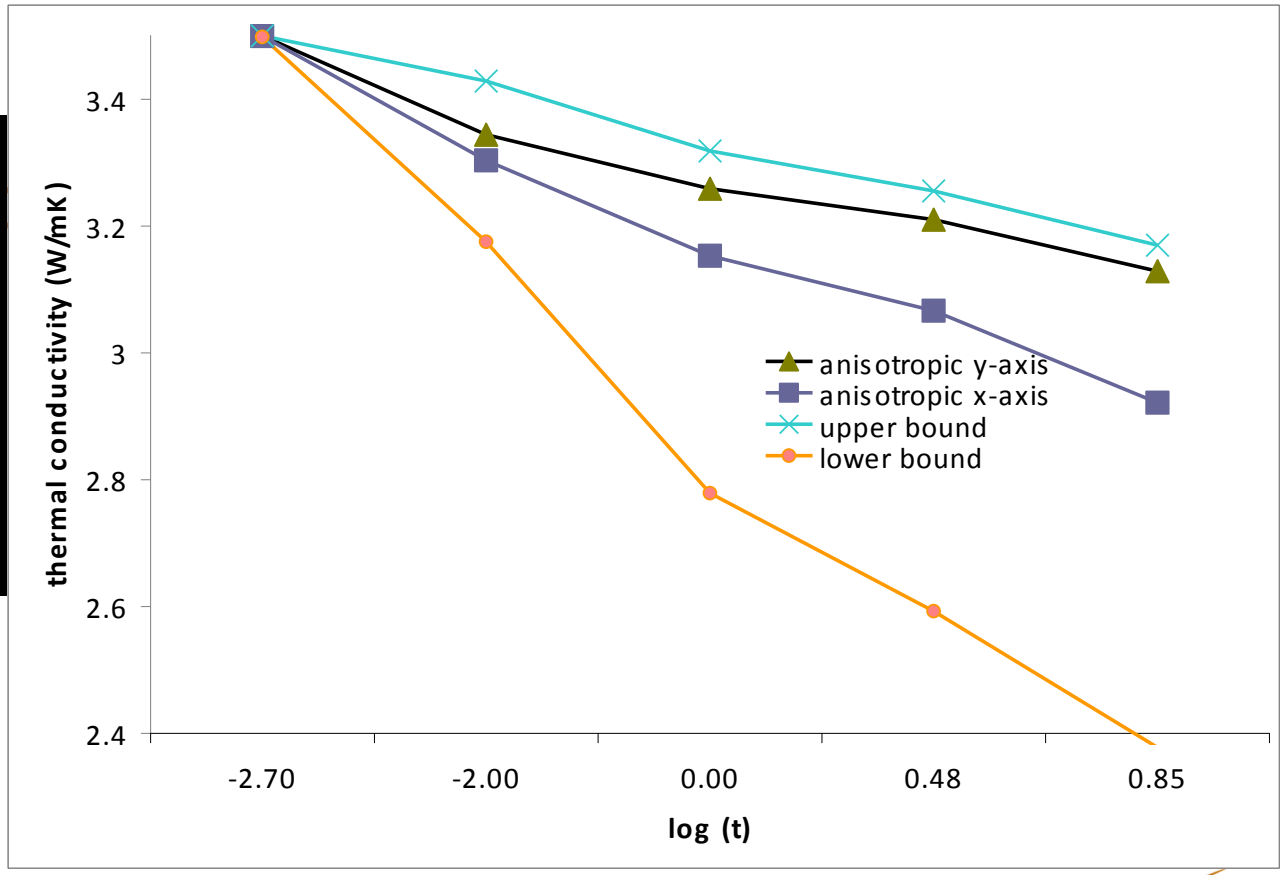


D.S. Li, et al. Modeling thermal conductivity evolution of nuclear materials under irradiation using multiscale approach. Journal of Nuclear Materials, submitted.

Example 2: Assessment of Upscaling Methodologies - From Phase field Model to Continuum Properties



UO2 example



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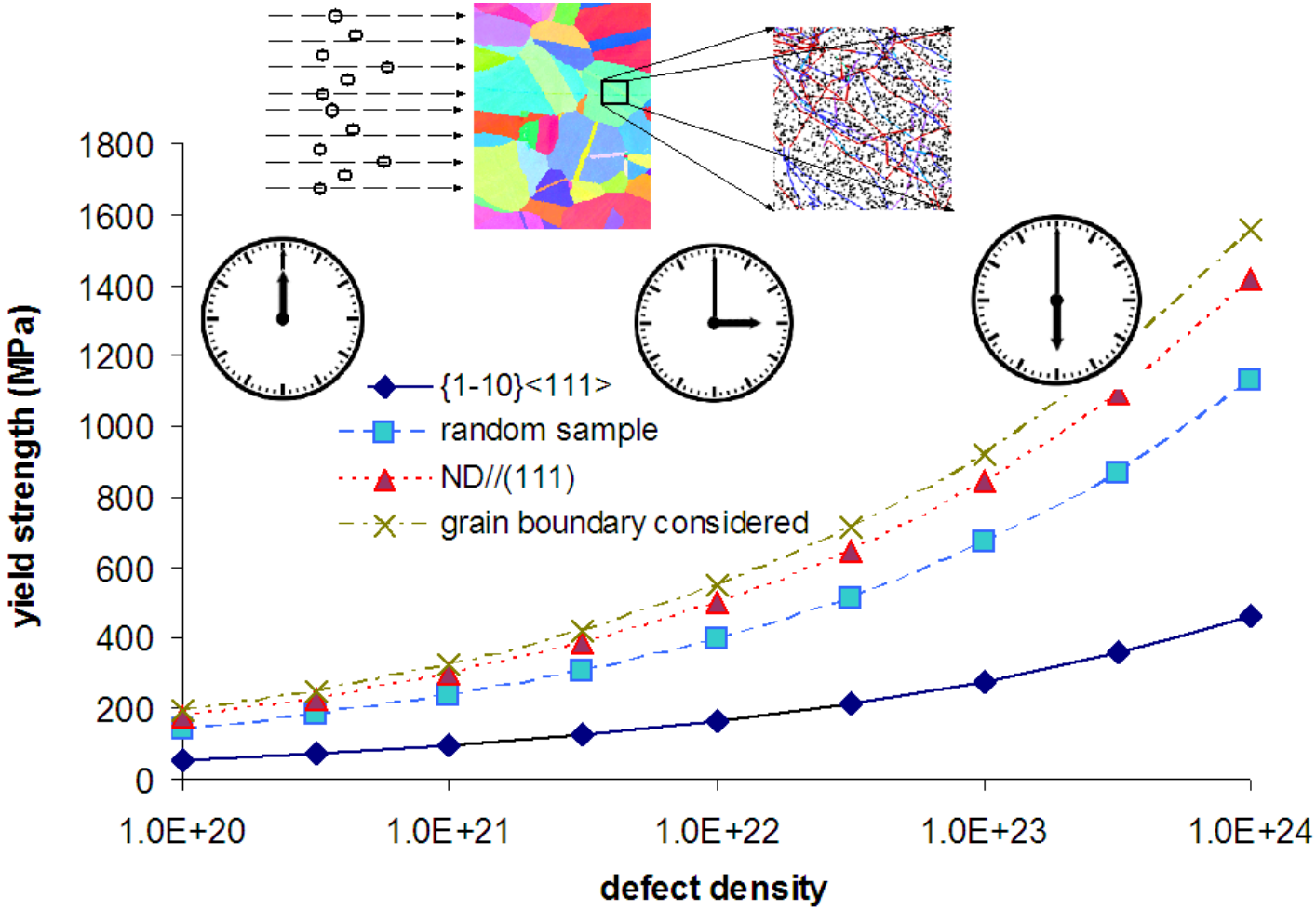
Strain Hardening
- Effect of Defect Density and Texture



D.S. Li et al. Composite Self-Consistent Model for Irradiated Steel: Effect of Defect Density and Grain Boundary,. Computational Materials Science, submitted.

Work Plans for FY11

- Phase field modeling:
  - Incorporation of thermodynamic and kinetic data predicted by lower length scale models
  - Calculation of long-range interaction energy
  - Improvement of computational efficiency – Collaboration with Fuels IPSC
- Scale bridging methodology:
  - Statistical model for reconstructing signature 3D heterogeneous multiphase microstructures
  - Constitutive level thermo-mechanical properties prediction for isotropic and anisotropic microstructures

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