INL/CON-22-68933-Revision-0

Phase-field modeling of radiation-induced segregation for multicomponent alloys

October 2022

Sourabh Bhagwan Kadambi, Daniel Schwen, Lingfeng He, Yongfeng Zhang

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Idaho National Laboratory

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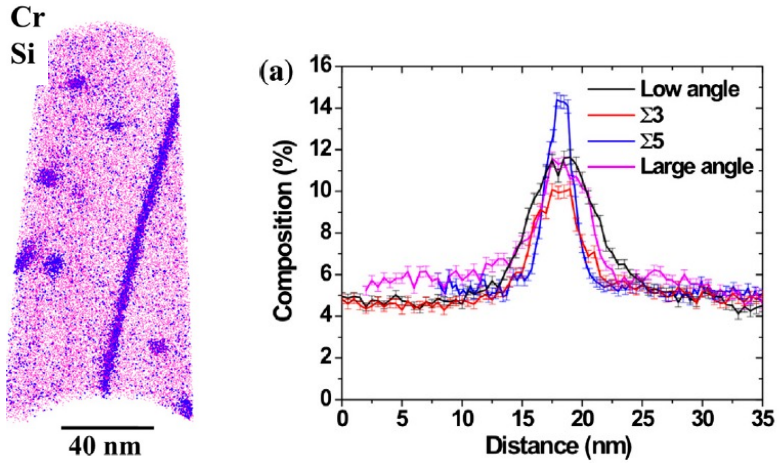
Idaho National Laboratory

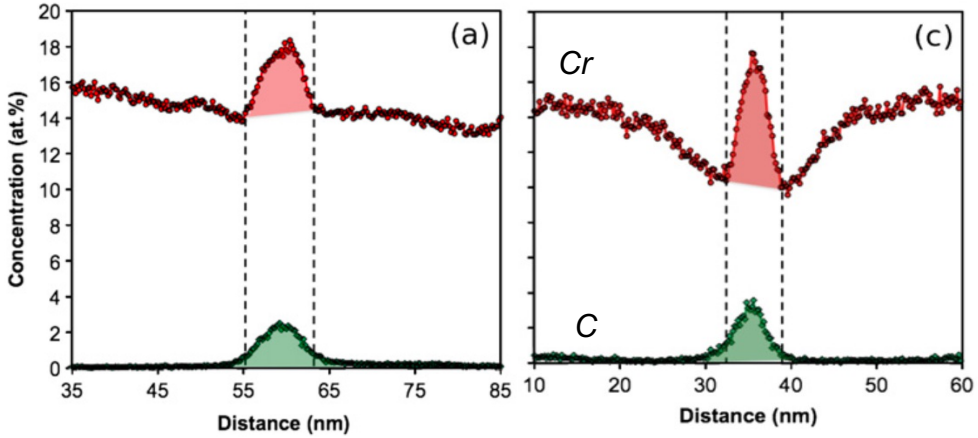
Radiation-induced composition redistribution (RIS)

Monotonous V-shaped RIS profiles [1]



APT image of segregation of neutron irradiated Fe-6at.% Cr (1.82 dpa, 290 C)

W-shaped RIS profile [2]



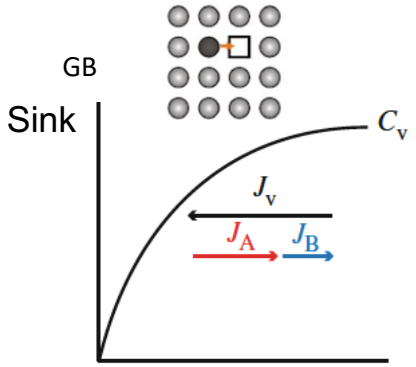
Concentration across GB in Fe-15at.%Cr (left) unirradiated,(right) ion irradiated to 8 dpa

- RIS has complex dependence on temperature, irradiation conditions and microstructural features
- Capability required: multicomponent alloys, non-ideal kinetics, non-ideal GB sink and other segregation mechanisms

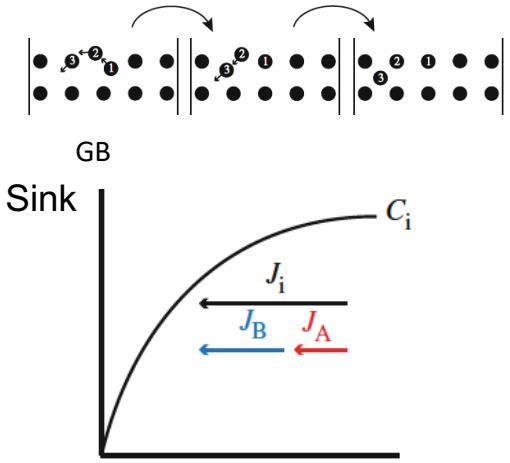
[1] Bachhav et al. , J. Nucl. Mater. 453 (2014)334 – 339
[2] Marquis et al. J. Nucl. Mater. 413 (2011) 1 – 4
[3] Ardell and Bellon, Curr. Opin. Solid State Mater Sci. 20(3) (2016) 115

Diffusion theory for RIS: atom-defect coupled transport

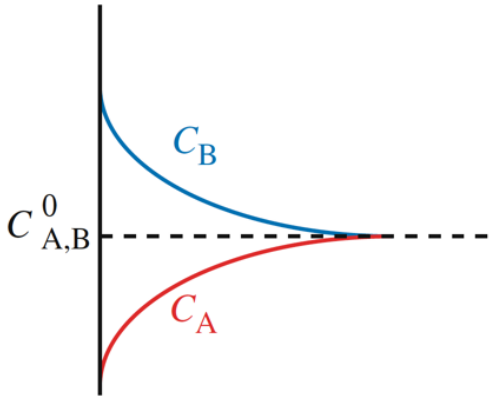
Atom-vacancy exchange mechanism



Self-interstitial dumbbell mechanism



Atomic redistribution



Enrichment/ depletion of atoms due to difference in partial fluxes

[1] Was, G. S. Fundamentals of Radiation Materials Science, Springer (2016)

Diffusion theory for RIS: atom-defect coupled transport

Partial fluxes for atoms A, B, C…

$$J_A^V = -L_{AA}^V \nabla(\mu_A - \mu_V) - L_{AB}^V \nabla(\mu_B - \mu_V) - \cdots$$

$$J_A^I = -L_{AA}^I \nabla(\mu_A + \mu_I) - L_{AB}^I \nabla(\mu_B + \mu_I) - \cdots$$

$$J_A = J_A^V + J_A^I$$

Total fluxes (reduced form B, C…)

$$J_B = -\tilde{L}_{BB} \nabla\tilde{\mu}_B - \tilde{L}_{BC} \nabla\tilde{\mu}_C \ldots - L_{BV} \nabla\mu_V - L_{BI} \nabla\mu_I \quad \text{(atoms)}$$

$$J_\nu = -\tilde{L}_{\nu B} \nabla\tilde{\mu}_B - \tilde{L}_{\nu C} \nabla\tilde{\mu}_C \ldots - L_{\nu\nu} \nabla\mu_\nu \quad \text{(defects)}$$

Chemical potential-based diffusion, rate theory

$$\chi_{BB} \frac{\partial\tilde{\mu}_B}{\partial t} + \chi_{BC} \frac{\partial\tilde{\mu}_C}{\partial t} + \cdots = -\nabla . J_B$$

$$\chi_{\nu\nu} \frac{\partial\mu_\nu}{\partial t} = -\nabla . J_\nu + \dot{P}_\nu - \dot{R}_{VI} - \dot{S}_\nu$$

Production | Recombination | Sink

Diffusion model for RIS:

- Full thermodynamic & kinetic coupling between atomic components and defects
- Solve evolution equation in terms of chemical potentials: required chemical susceptibilities
- MOOSE: open-source, finite element, C++

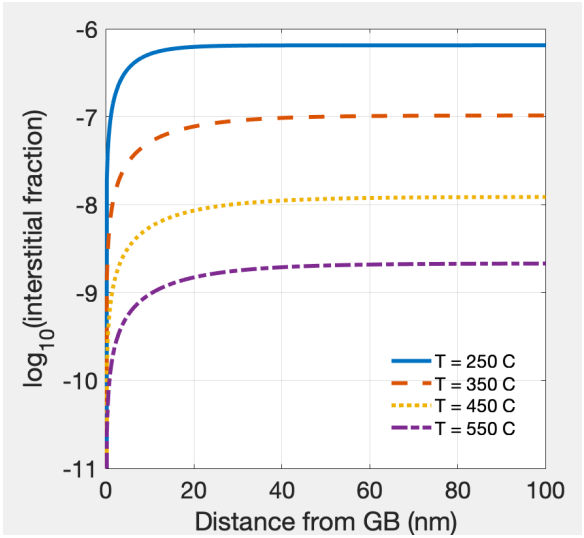
[1] Wolfer, W.G., J. Nucl. Mat., 114 (1983) 292−304
[2] Aagesen et al., Comp. Mat. Sci. 161 (2019) 35−45

MOOSE

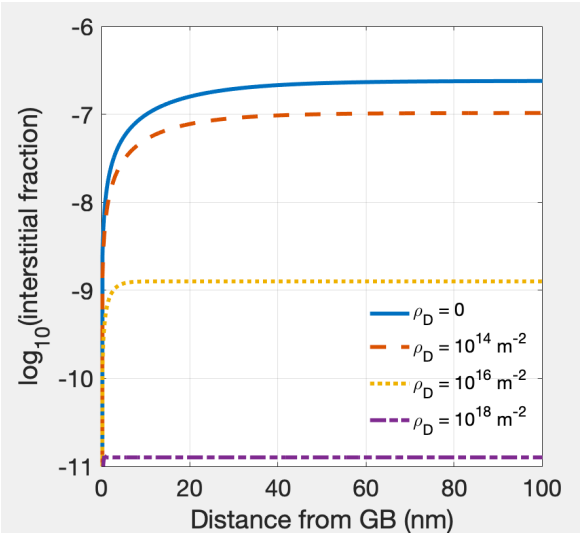
Verification of implementation of MIK model for Fe-Cr-Ni

MOOSE

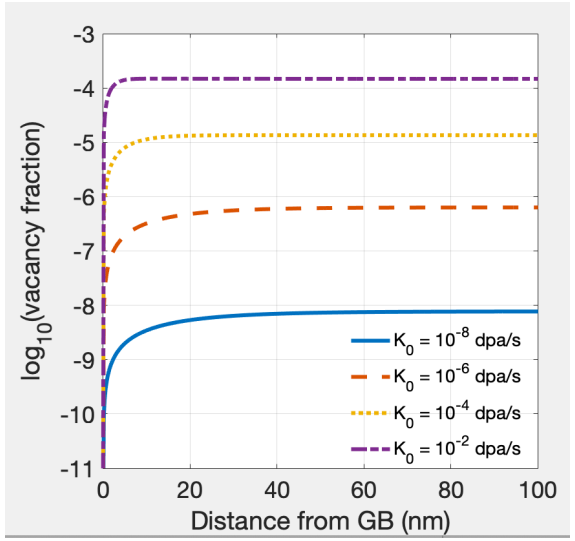
Temperature



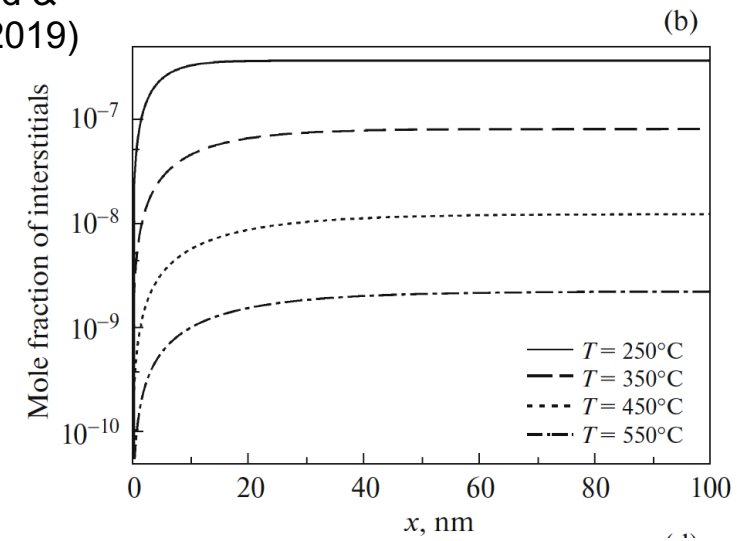
Dislocation density

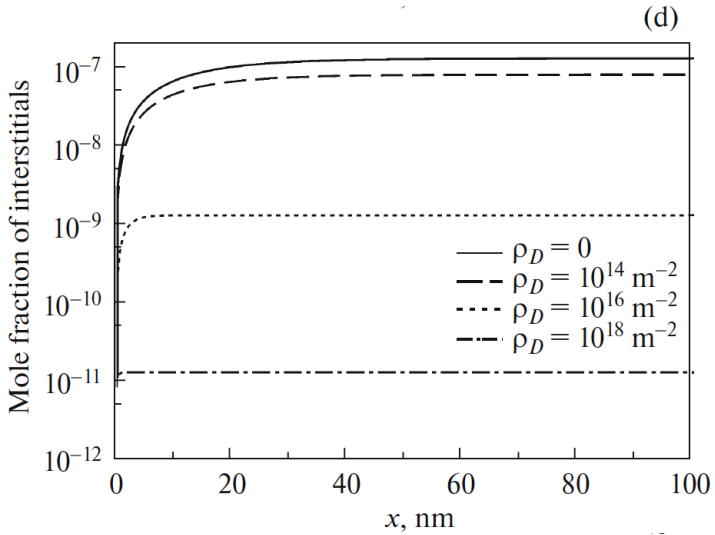


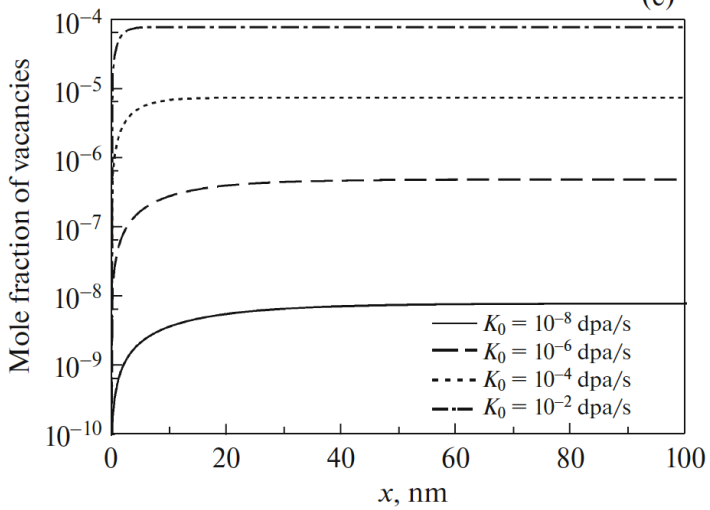
Defect production rate



Skorokhod & Koropov (2019)







Parameterization for FCC NiCrFe: thermodynamics & kinetics

- CALPHAD free energy is approximated using Taylor expansion about nominal composition

$$\begin{pmatrix} \tilde{\chi}_{BB} & \tilde{\chi}_{BC} \\ \tilde{\chi}_{CB} & \tilde{\chi}_{CC} \end{pmatrix} = \begin{pmatrix} \widetilde{\Theta}_{BB} & \widetilde{\Theta}_{BC} \\ \widetilde{\Theta}_{CB} & \widetilde{\Theta}_{CC} \end{pmatrix}^{-1}, \qquad \chi_{vv} = \Theta_{vv}^{-1}$$

- Ideal solution free energy for point defects

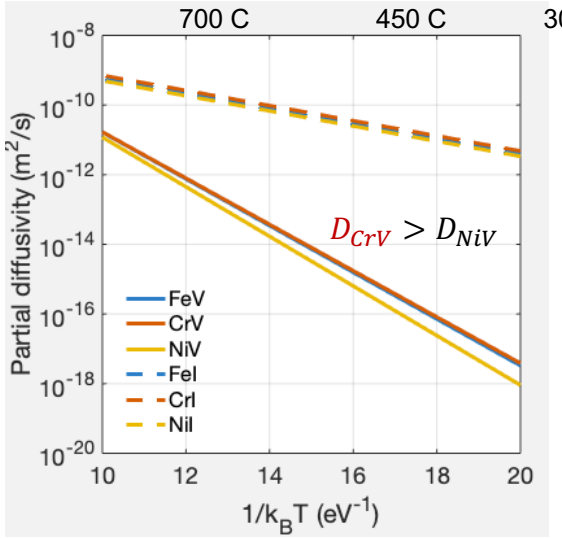
- Vacancy-based transport: from CALPHAD mobility data [1]

$$D_{iV} = \frac{D_{iV}^{*}}{x_V^{eq}} = P_i \exp\left(-\frac{Q_i - E_V^f}{k_B T}\right)$$

- Interstitial-based transport: equal migration barrier from MIK model; binding energies used as fitting parameters [2]

$$D_{iI} = \frac{1}{6}\lambda_I^2 z f_I \omega_{iI} \exp\left(-\frac{E_I^m}{k_B T}\right)\beta_i$$

Partial diffusivities for Ni-18Cr-9Fe



Onsager transport from partial diffusivities using Manning-type analytic relation

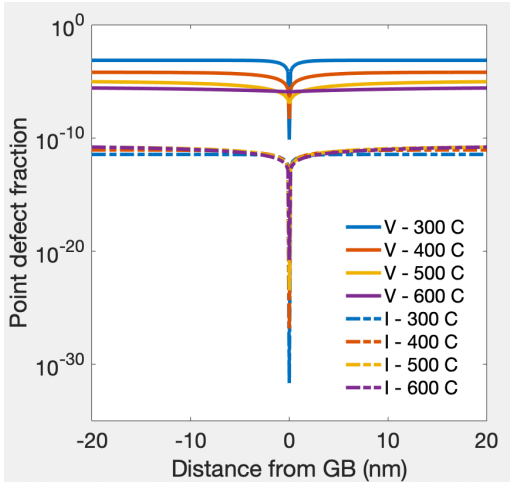
$$L^V = \begin{pmatrix} L_{AA}^V & L_{AB}^V & L_{AC}^V \\ L_{AB}^V & L_{BB}^V & L_{BC}^V \\ L_{AC}^V & L_{AB}^V & L_{CC}^V \end{pmatrix} \qquad \frac{L_{BV}}{L_{BB}^V} = -1$$

Ideal kinetics

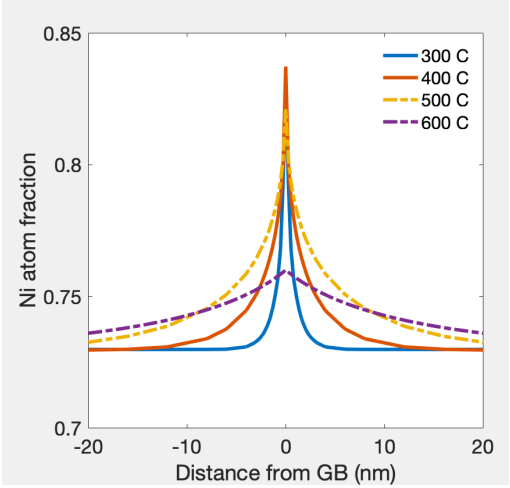
[1] Yang et al., J. Nucl. Mater. 473 (2016) 35 – 53
[2] Allen et al., J. Nucl. Mater. 255 (1998)44 – 58 (MIK model)

Verification of RIS implementation for FCC Ni-9Fe-18Cr

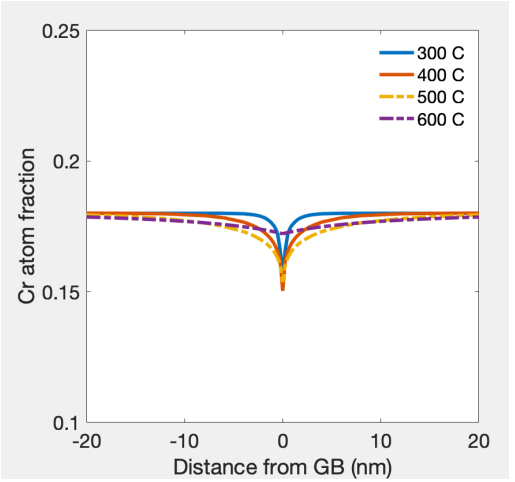
Point defect profile



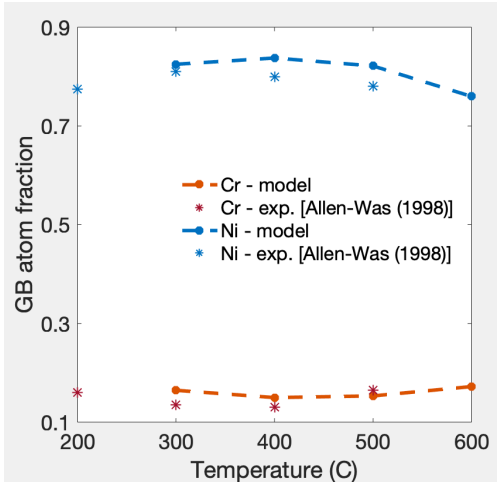
Ni enrichment



Cr depletion



Comparison with experimental data



$K_o = 7\times10^{-6}\ dpa/s$

$S_{gb} = 10^9 1/\mathrm{s}$

$\rho_D = 10^{14} m^{-2}$

- Using parameters of Yang (2016), good match with existing models and experimental data of Allen-Was (1998)

General assumptions:

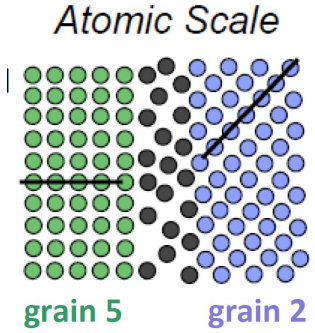
- Ideal kinetics, ideal GB sink, only RIS mechanism
- Simple 1D geometries: planar, stationary GB

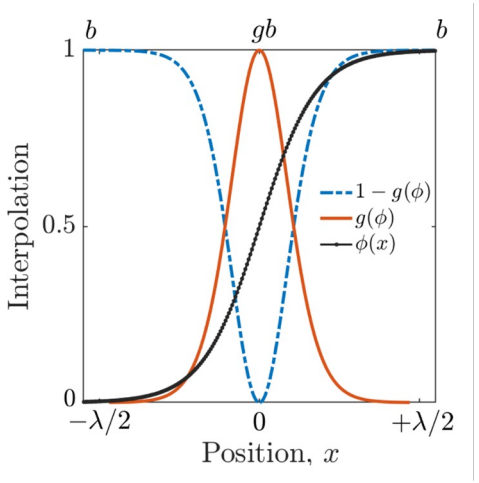
Phase-field method can relax these assumptions

[1] Yang et al. , J. Nucl. Mater. 473 (2016) 35 – 53
[2] Allen et al. , J. Nucl. Mater. 255 (1998)44 – 58 (MIK model)
[3] Skorokhod & Koropov, Phys. of the Solid State, 61 (2019) 2269 – 2276

RIS coupled with grand potential phase-field model

MOOSE





Grand potential functional

$$\Psi = \int_V \left\{ \omega\left(\tilde{\mu}_B, \tilde{\mu}_C, \mu_V, \mu_I, \eta\right) + W_\eta g(\eta) + \frac{\kappa}{2}|\nabla\eta|^2 \right\} dV$$

Chemical potential evolution

$$\chi \frac{\partial \tilde{\mu}}{\partial t} = \nabla.\left(L\nabla\tilde{\mu}\right) - \frac{\partial x}{\partial \eta}\frac{\partial \eta}{\partial t} + P + S_{gb} g(\eta)$$

Phase-dependent local atomic concentration

$$x(\tilde{\mu}, \eta) = x^b(\tilde{\mu})[1 - g(\eta)] + x^{gb}(\tilde{\mu}) g(\eta)$$

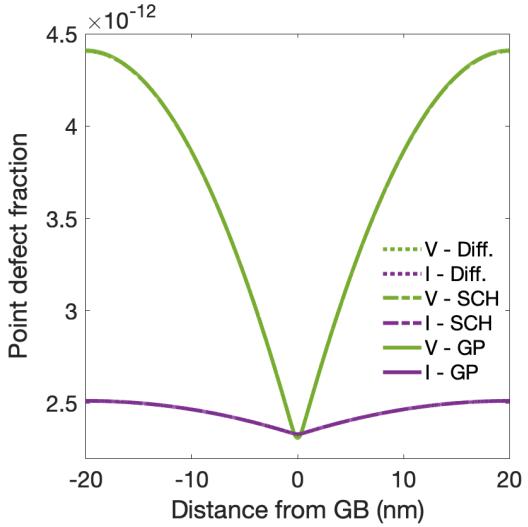
Allen-Cahn for η evolution

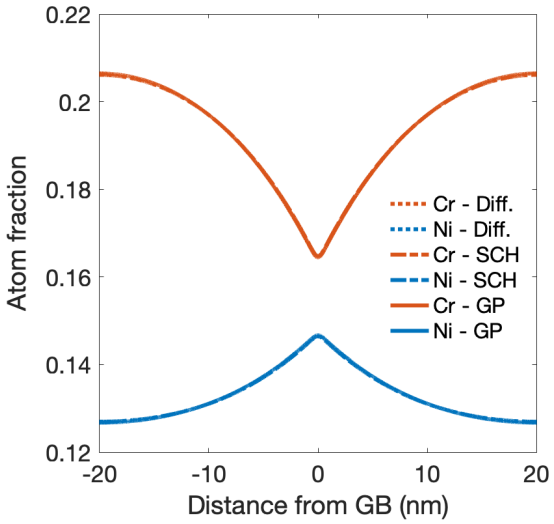
- Potential and phase-dependent kinetics: $L(\tilde{\mu}_B, \tilde{\mu}_C, \mu_V, \mu_I, \eta)$
- Phase-dependent susceptibilities: $\tilde{\chi}\left(\mu_B, \mu_C, \eta\right), \chi_V(\mu_V), \chi_I(\mu_I)$

[1] MOOSE workshop, PhaseField module, INL
[2] Aagesen et al., Phys. Rev. E 98 (2018) 023309

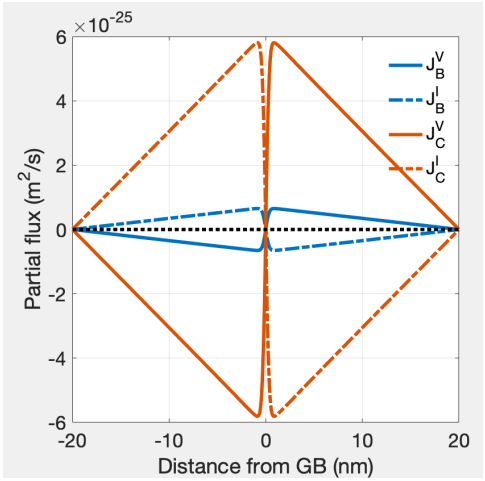
Grand potential phase-field model: cross-verification

Comparing different schemes



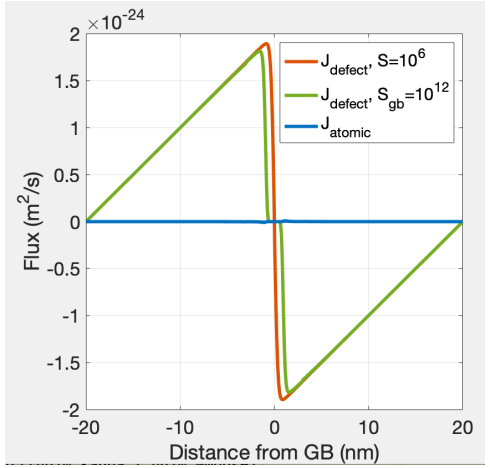


Steady-state fluxes from phase-field model



$J_B^V + J_B^I = 0$

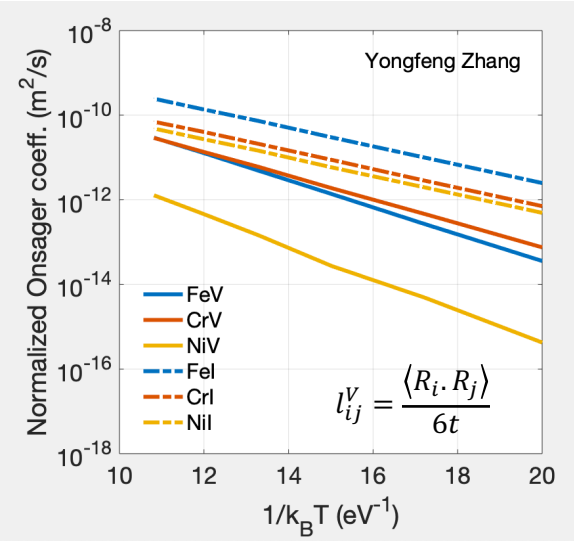
$J_C^V + J_C^I = 0$



$J_V = J_I$

$J_A + J_B + J_C = 0$

Onsager coefficients from AKMC [1]



Fe-19Cr-13Ni (at.%)

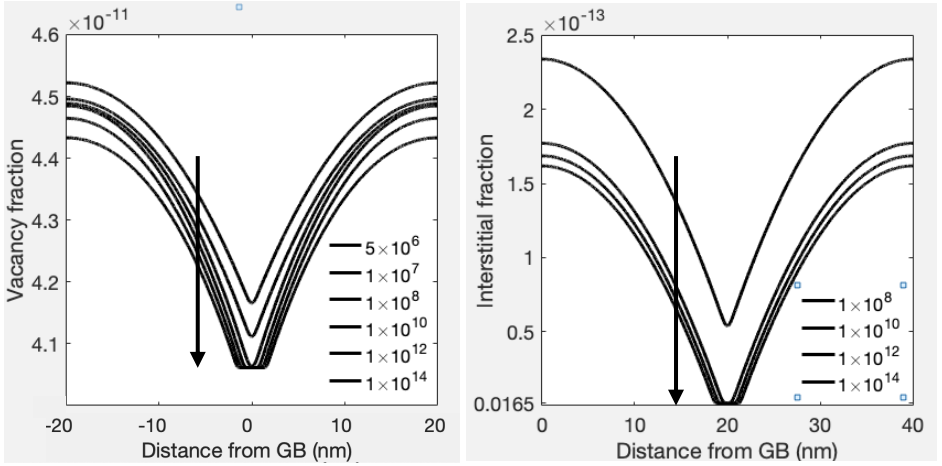
Temperature: 873 K
Source: 1×10^{-7} dpa/s
Sink strength: 2×10^{6} 1/s

Onsager coefficients from AKMC calculations

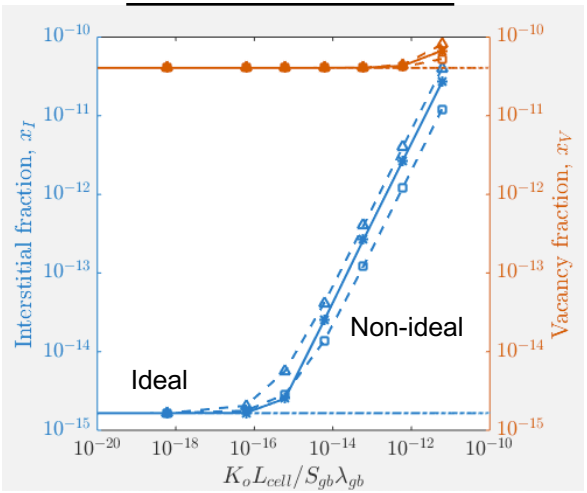
[1]Atomistic data from Yongfeng Zhang, UW – Madison

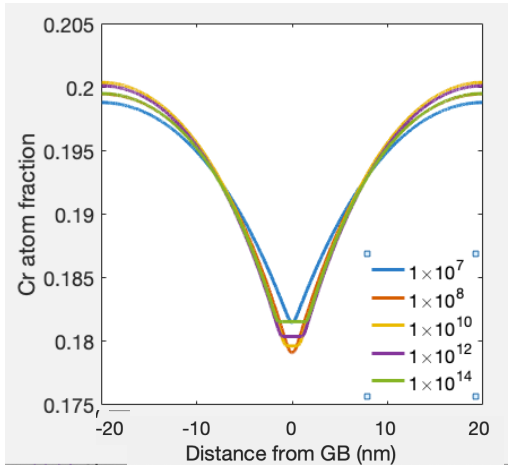
Effect of grain boundary sink coefficient S_{gb} on RIS

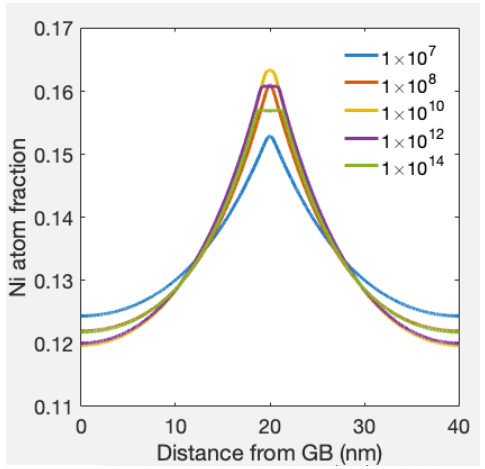
Profiles across GB

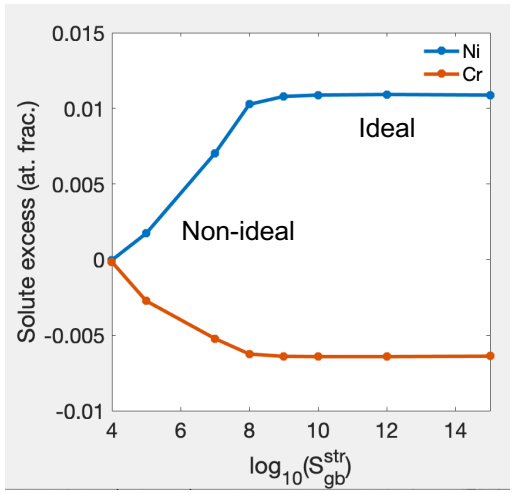


GB concentration









- Sink absorption rate: $S_{gb}(\rho,\theta)\left(x_v - x_v^{ideal}\right)g(\eta)$
- Verified simulation results with analytic solution derivable for Dirac/step-function

$$x_v^{ss} = x_v^{ideal} + K_o L_{cell}/S_{gb}\lambda_{gb}$$

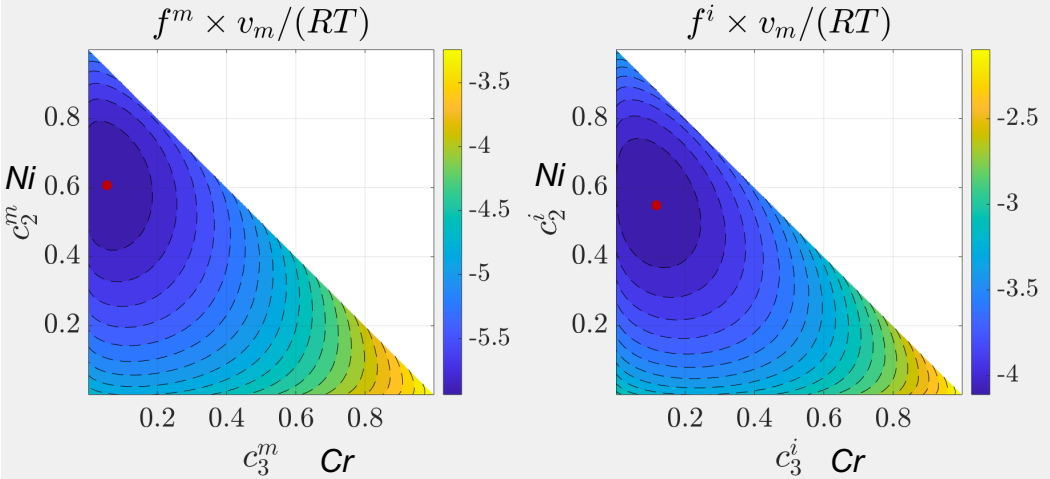
- Gibbs solute excess at GB with reference to bulk: better metric for RIS quantification

[1] Duh et al. , J. Nucl. Mater. 294 (2001) 267 – 273
[2] Nichols, J. Nucl. Mater. 75 (1978) 32 – 41

RIS in the presence of thermodynamic segregation for FeCrNi

Bulk free energy

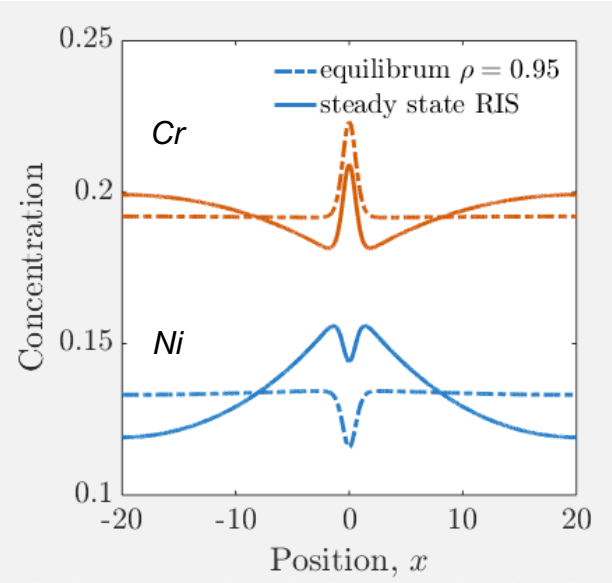
Grain boundary free energy



Bulk $\rho = 1$

Relative GB atomic density $\rho = 0.95$

W-shaped profile ≫ 10 dpa



$K_o = 10^{-7}\ dpa/s$

$S_{gb} = 10^6 1/s$

$T = 500$ C

$$f_{CALPHAD} = G_A(\rho)x_A + G_B(\rho)x_B + \cdots + L_{AB}(x,T,\rho)x_A x_B + \cdots - T\Delta S_{mix}$$

- Simple model to provide modified free energy landscape at grain boundary[2] due to atomic structure difference (can include GB-characteristic E_A, E_B, L_{AB} parameters)
- Successfully predicts steady-state W- or M-shaped profiles!

[1] Kamachali, R. D., RSC Adv. 10 (2020) 26728
[2] Kim et al., Acta Mat. 112 (2016) 150 – 160

Transient W-shape due to strong non-ideality in kinetics

Strong cross-coupling in kinetic transport
AKMC calculation for NiAlCr [1]

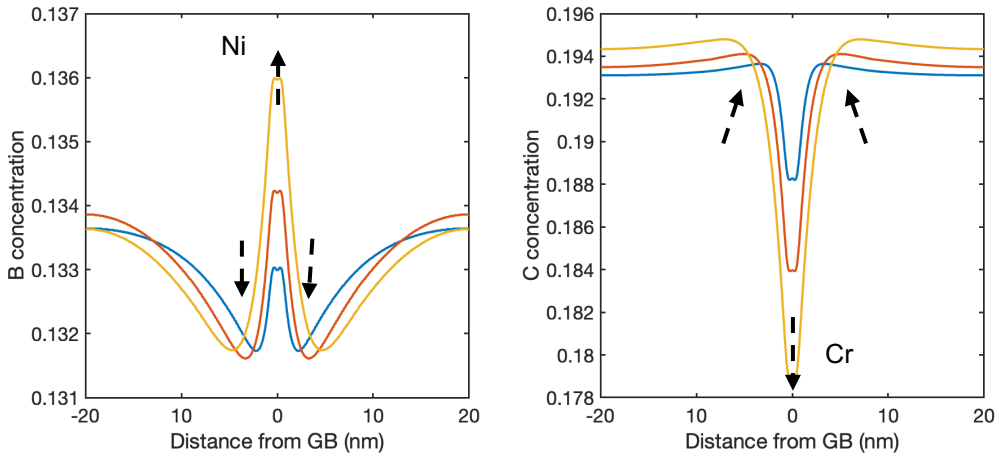
$$L = 10^{-24} \begin{pmatrix} 149 \pm 9 & 207 \pm 11 & 118 \pm 6 \\ - & 495 \pm 19 & 189 \pm 9 \\ - & - & 105 \pm 6 \end{pmatrix} (\mathrm{m^2\,s^{-1}})$$

$$\begin{pmatrix} NiNi & NiAl & NiCr \\ AlNi & AlAl & AlCr \\ CrNi & CrAl & CrCr \end{pmatrix}$$

$$\frac{L_{NiV}}{L^V_{NiNi}} = \frac{-(L^V_{NiNi} + L^V_{NiAl} + L^V_{NiCr})}{L^V_{NiNi}} < -1$$

Non-ideal kinetics for Ni and Cr

Effect of non-ideal kinetics on transient profiles
(irradiation dose ≪ 0.1 dpa)



- System with strong negative coupling of solute with vacancy (non-dilute limit) [1]
- Potential flip in sign of RIS or change in extent of RIS
- Long diffusion tails in near-GB region can result in W or M-shaped transient profiles
- Full kinetic coupling: can model solute drag mechanism for ratio > -1

[1] Mao et al. Acta Mat. 60 (2012)1871 – 1888
[2] Senninger et al., Acta Mat. 103 (2016) 1 – 11

Future scope

- A phase-field RIS model is being developed with capability of modeling non-ideal kinetics, equilibrium GB segregation and non-ideal GB sink
- Better predictions are possible with improved input/ coupling with atomistic calculations (especially interstitials)
- Scope for systematic investigation of RIS for different grain boundary characters/ misorientations, validation of W-shaped profiles
- Scope for extension to multi-order parameter polycrystalline phase-field model
- Coupling with cluster dynamics-type model can provide physically-based fluxes to GB can improve RIS predictions