Microstructural Evolution in Interdiffusion Zone and Its Effect on Diffusion Path

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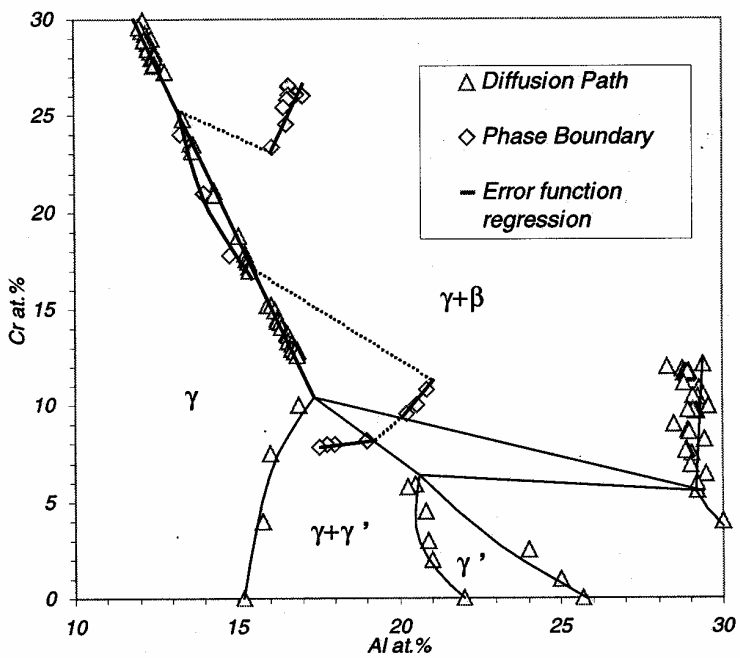
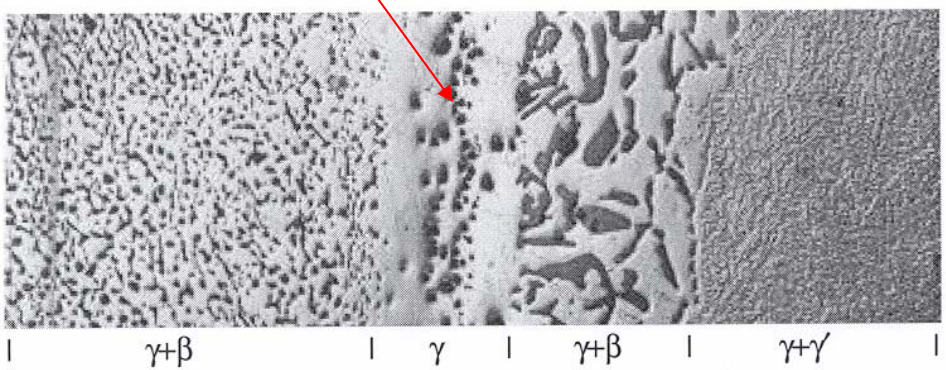
Experimental Observation of Interdiffusion Microstructure and Diffusion Path

$\gamma+\beta<\gamma>\gamma+\beta>\gamma+\gamma'$



(c) Diffusion path of diffusion couple

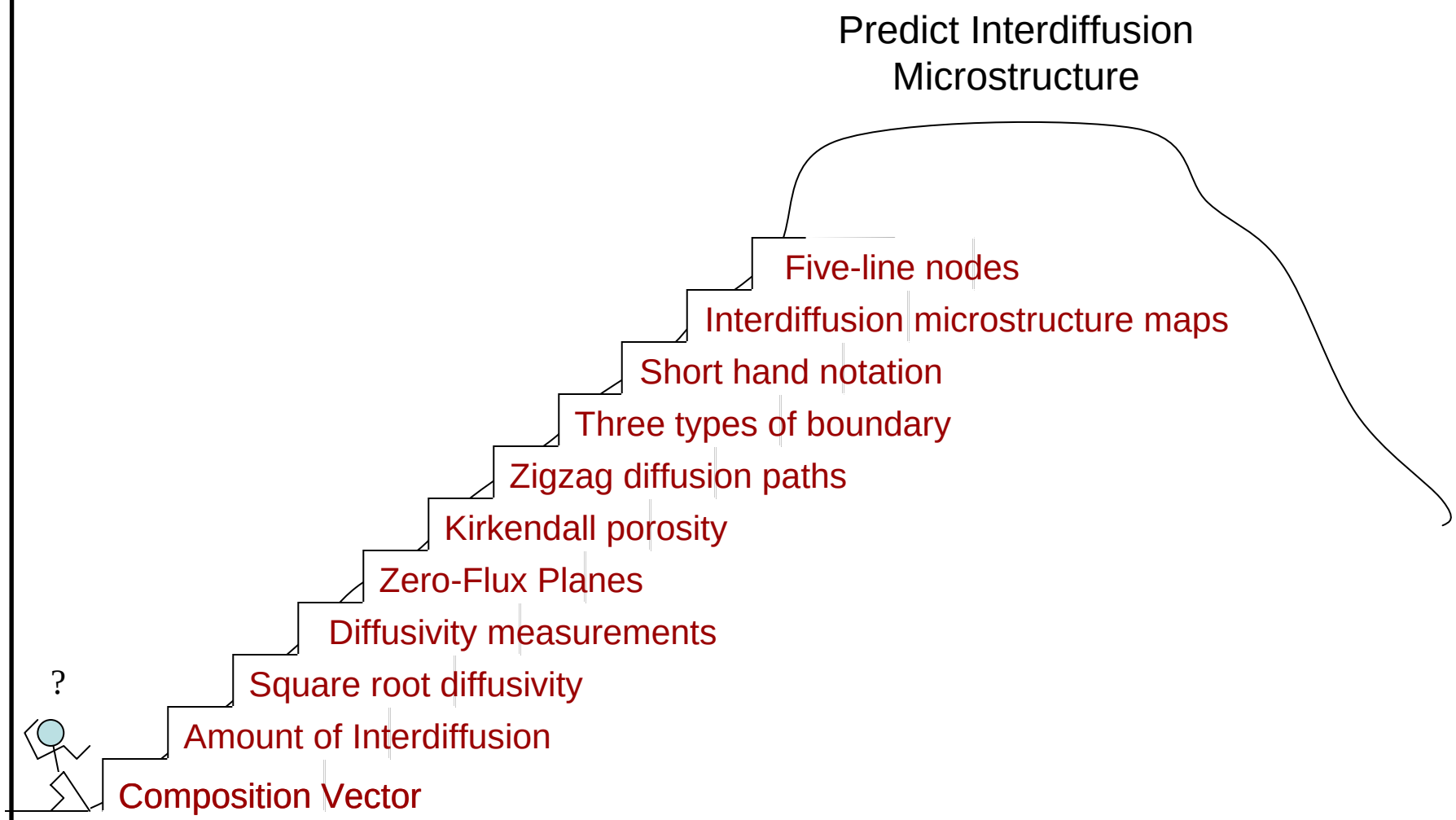
Kirkendall porosity



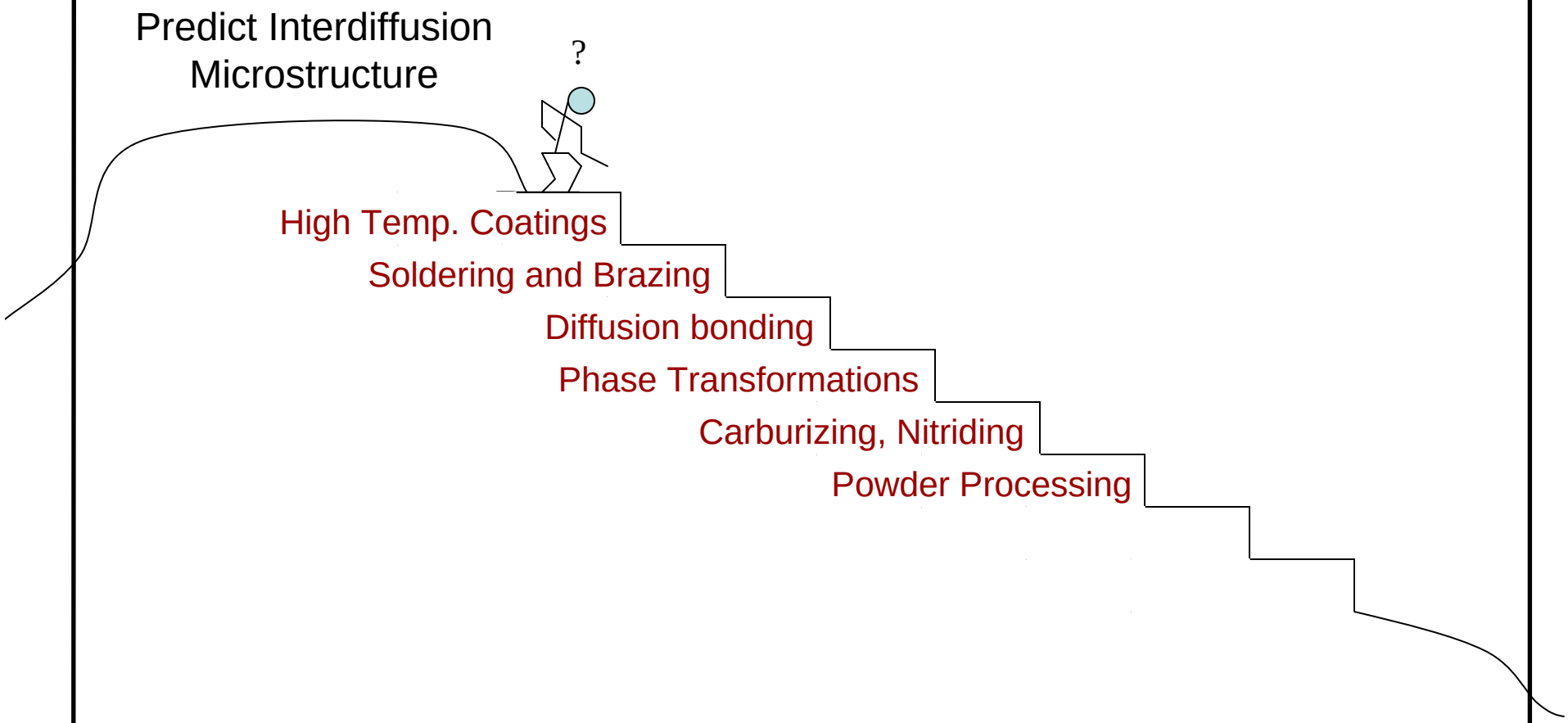
(d) Microstructure of diffusion couple

Xin Qiao. M.S. Thesis. University of Connecticut. 1998

The Multicomponent Mountain



The Other Side of the Multicomponent Mountain



Climbing the Multicomponent Mountain with John

Coupling interdiffusion with microtructural evolution:

- Effect of two-phase microstructure on interdiffusion and diffusion path
- Interdiffusion induced phase and microstructure instabilities
- Effect of concentration gradient on nucleation, growth and coarsening
- Effect of phase transformation on interdiffusion
- Roles of coherency/thermal stress on interdiffusion and phase transformation

Predict Interdiffusion Microstructure

Phase Field

Five-line nodes

Interdiffusion microstructure maps

Short hand notation

Three types of boundary

Zigzag diffusion paths

Kirkendall porosity

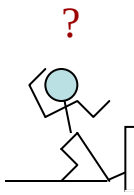
Zero-Flux Planes

Diffusivity measurements

Square root diffusivity

Amount of Interdiffusion

Composition Vector

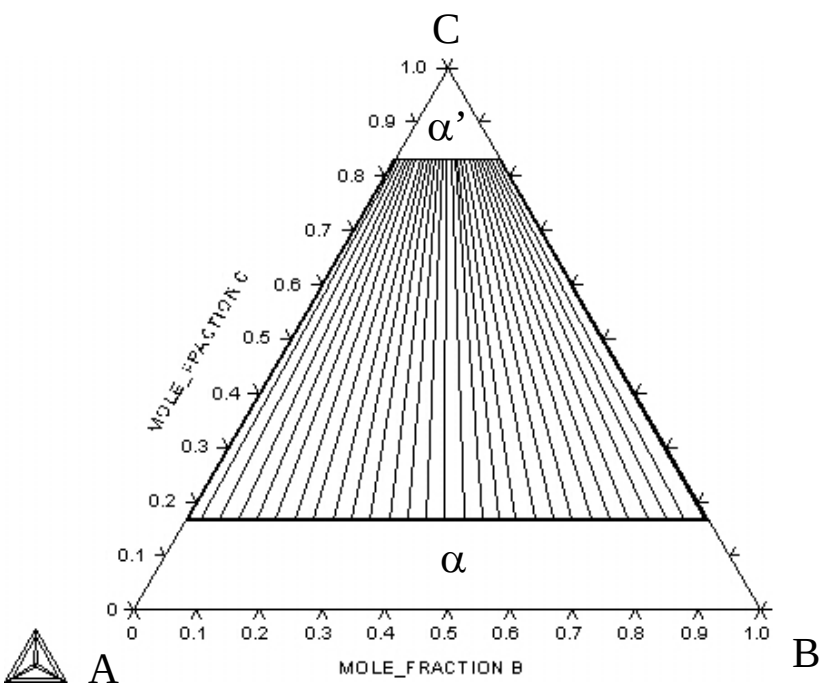


- One-dimensional diffusion in a common matrix phase
- Precipitates are treated as stationary point sources or sinks of solute
- Mutual interactions between microstructure and interdiffusion and corresponding effects on diffusion path and microstructural evolution are ignored

Simple Model System

Free energy model

$$G_m = RT\left(X_A \ln X_A + X_B \ln X_B + X_C \ln X_c\right) + I\left(X_A X_C + X_B X_C\right)$$



- Elements A and B form ideal solution while elements A and C or B and C form regular solutions

Wu et. al. Acta mater. 2001;49:3401
Wu et. al. Acta mater, 2004; 52:1917

Phase Field Equations

$$\left.\begin{aligned}\frac{\partial X_B}{\partial t} &= \nabla\left[M_{11}\nabla\left(\mu_B-\mu_A\right)\right]+\nabla\left[M_{12}\nabla\left(\mu_C-\mu_A\right)\right]\\ \frac{\partial X_C}{\partial t} &= \nabla\left[M_{21}\nabla\left(\mu_B-\mu_A\right)\right]+\nabla\left[M_{22}\nabla\left(\mu_C-\mu_A\right)\right]\end{aligned}\right\}$$

Diffusion equations

$$\left.\begin{aligned}\mu_B-\mu_A &= \mu_B^B-\mu_A^B-2\kappa_{11}\nabla^2 X_B-2\kappa_{12}\nabla^2 X_C\\ \mu_C-\mu_A &= \mu_C^B-\mu_A^B-2\kappa_{21}\nabla^2 X_B-2\kappa_{22}\nabla^2 X_C\end{aligned}\right\}$$

Gradient thermodynamics

$$\left.\begin{aligned}M_{11} &= \rho X_B\left[\left(1-X_B\right)^2\beta_B+X_B X_C\beta_C+X_B X_A\beta_A\right]\\ M_{12}=M_{21} &= \rho X_B X_C\left[-\left(1-X_B\right)\beta_B-\left(1-X_C\right)\beta_C+X_A\beta_A\right]\\ M_{22} &= \rho X_C\left[X_B X_C\beta_B+\left(1-X_C\right)^2\beta_C+X_C X_A\beta_A\right]\end{aligned}\right\}$$

Kinetics parameters

M_{ij} - chemical mobilities

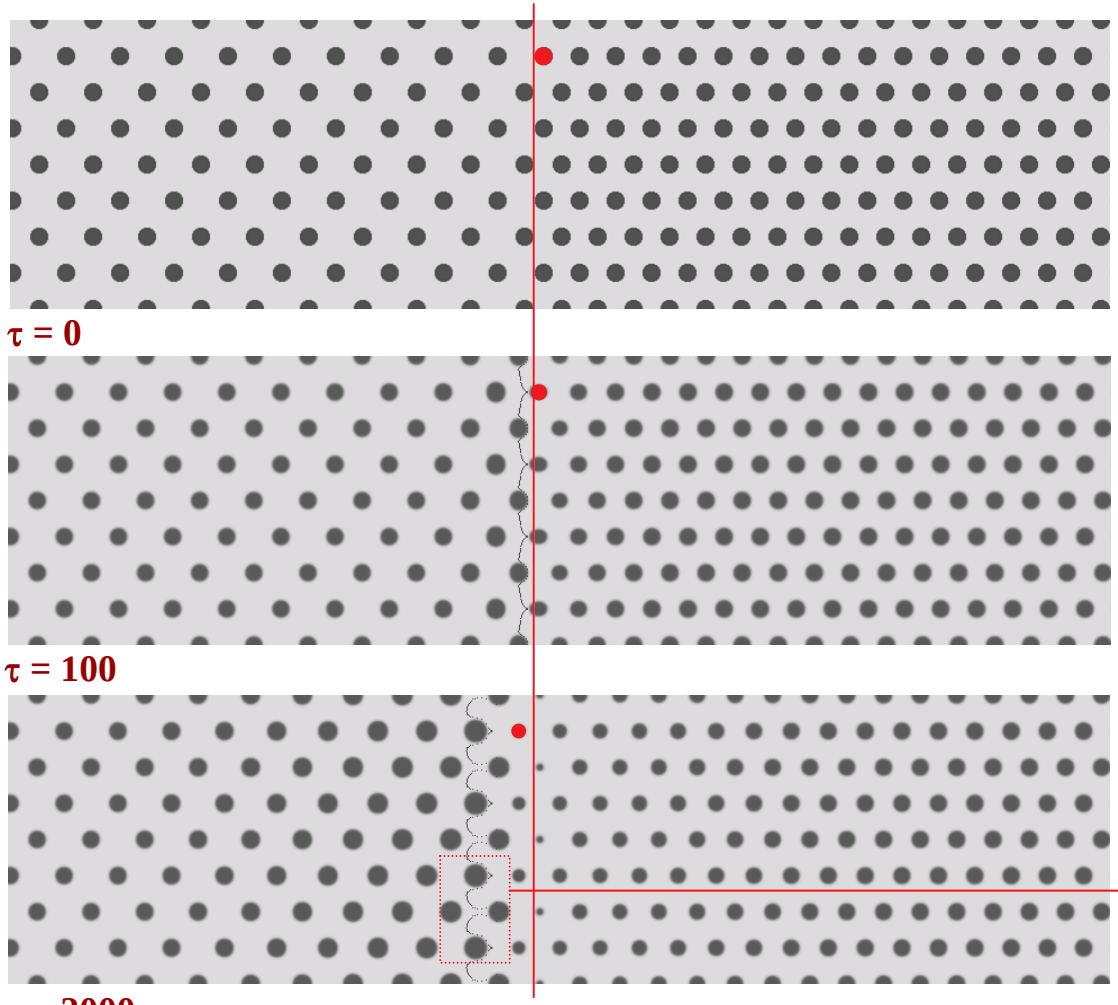
κ_{ij} - gradient coefficients

β_I - atomic mobilities

ρ - molar density

Wu et. al. Acta mater. 2001;49:3401
Wu et. al. Acta mater, 2004; 52:1917

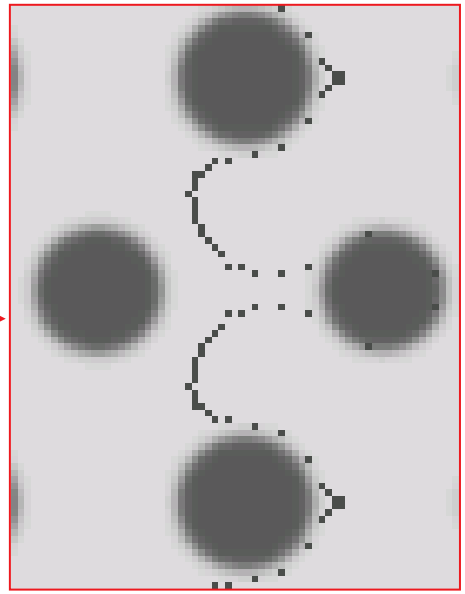
Interaction between Microstructure and Interdiffusion – Type 0 boundary



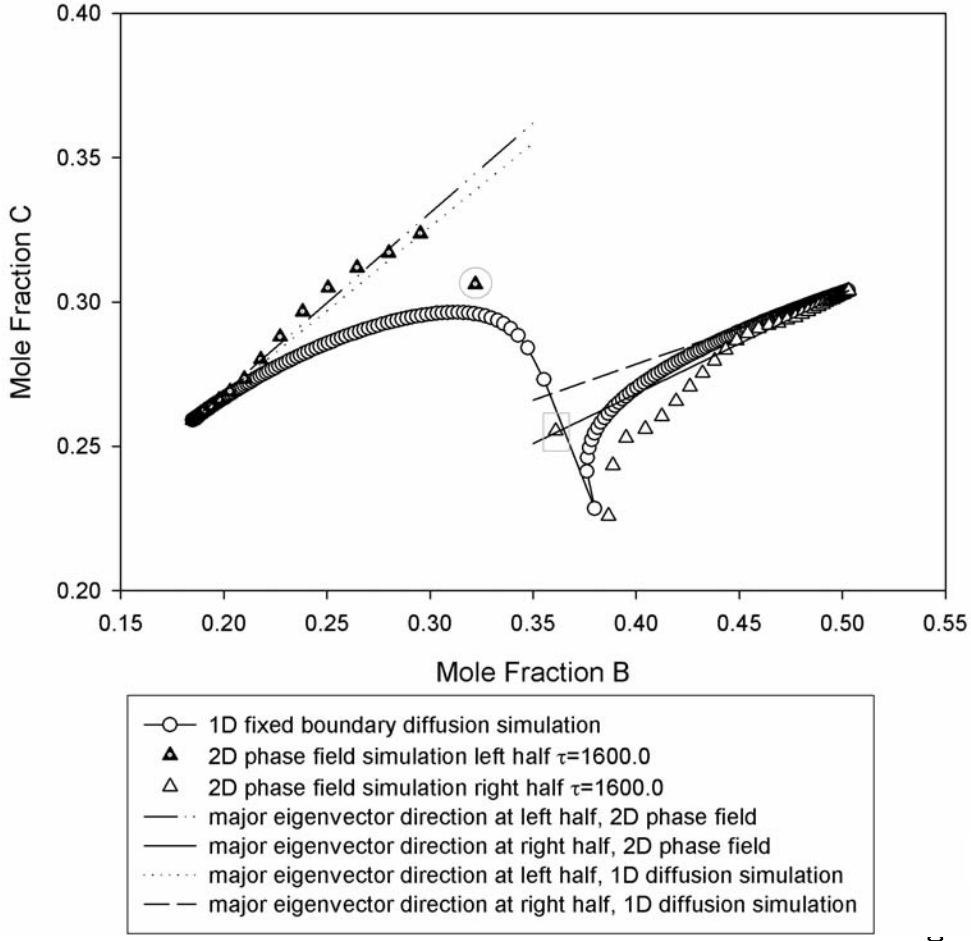
β_B=1.0 β_C=5.0 β_A=10.0

4608x64 size simulation, 1024x256 size output

- Ppt and Type 0 boundary migrate as a results of Kirkendall effect
- Type 0 boundary becomes diffuse
- Kirkendall markers move along curved path and marker plane bends around precipitates
- Diffusion path differs significantly from 1D calcul.

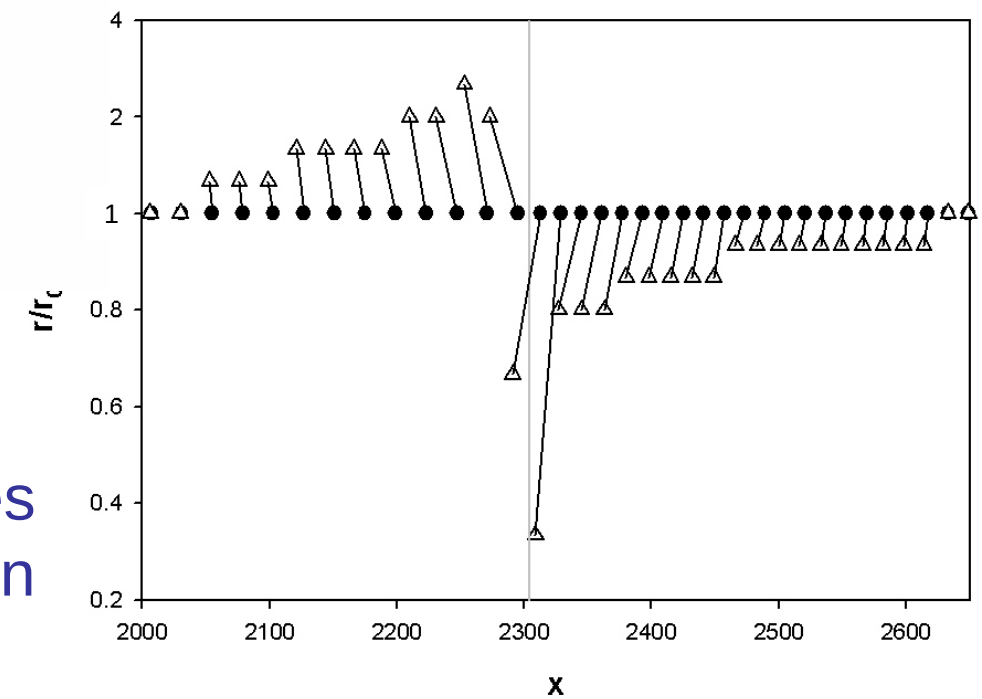


Diffusion path: comparison with 1D simulation



Wu et. al. Acta mater. 2001;49:3401
Wu et. al. Acta mater, 2004; 52:1917

Size and position changes during interdiffusion



Real Alloy System: Ni-Al-Cr

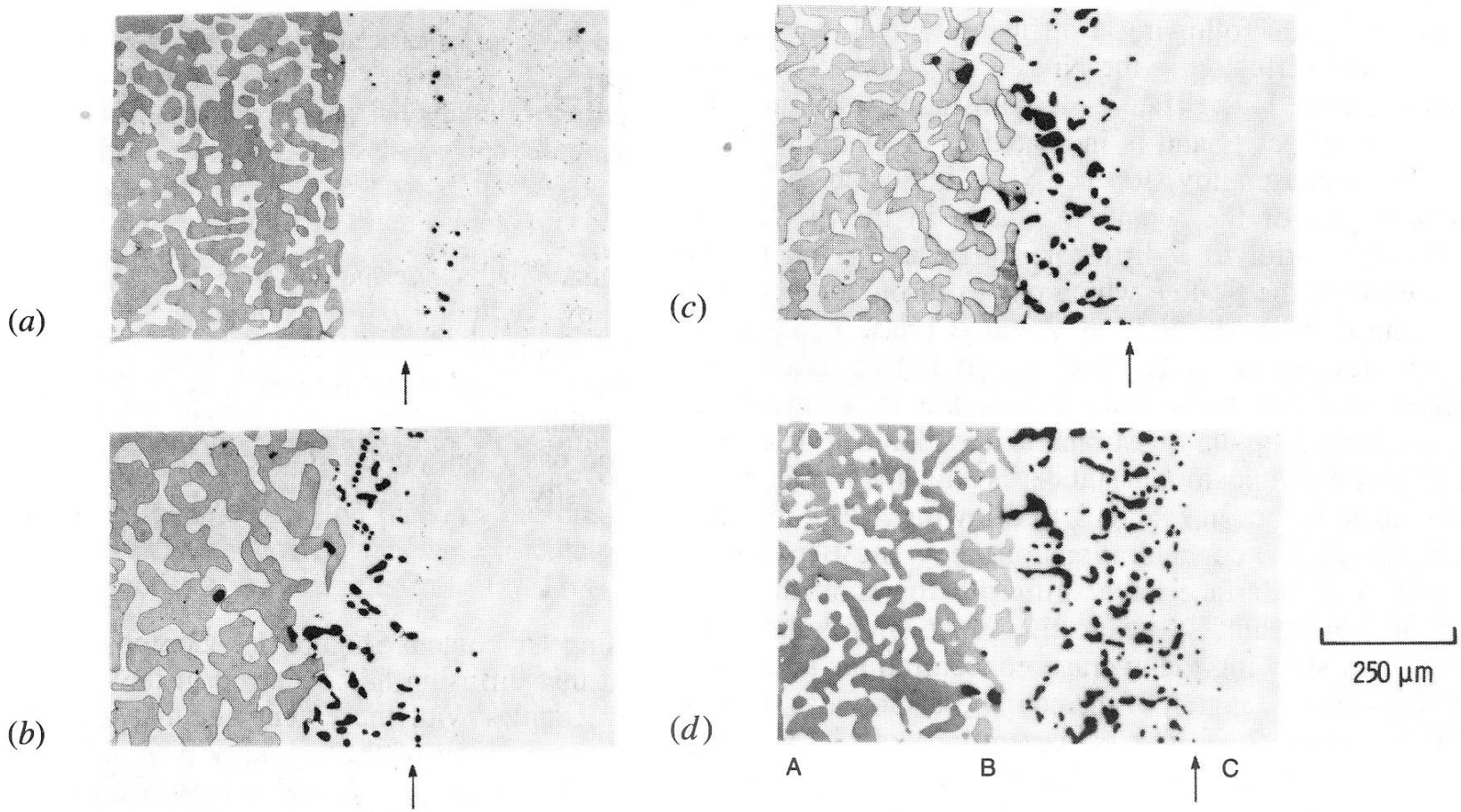


Fig. 1—Microstructures of four $\gamma/\gamma + \beta$, Ni-Cr-Al diffusion couples after 100 h at 1200 °C. Arrows indicate original couple interface. Dark phase is β. Kirkendall porosity (black) is located in the diffusion zone (see Ref. 7). (*a*) Couple Ni-12.7Cr-24.0Al/Ni-45.5Cr, β recession = 120 μm. (*b*) Couple Ni-12.7Cr-24.0Al/Ni-35.2Cr, β recession = 140 μm. (*c*) Couple Ni-12.7Cr-24.0Al/Ni-25.0Cr, β recession = 205 μm. (*d*) Couple Ni-12.7Cr-24.0Al/Ni, β recession = 320 μm.

Exp. Observation by Nesbitt and Heckel in Met Trans. A (1986)18A: 2087-2094

Linking to Thermo. and Kinetic Databases and Atomistic Calculations

Database development

Experimental Data

Atomistic Calculations

Optimizer

Mobility Database

DICTRA

TD Database

Thermo_Calc or PANDAT

Kinetic description

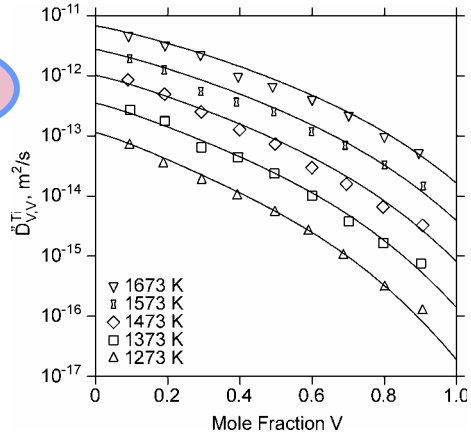
Thermodynamic description

Interface Properties

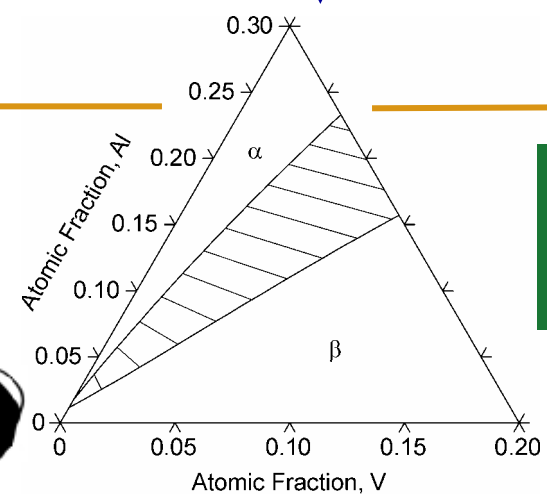
Elastic Properties

γ-surfaces

Other properties



Chemical Diffusivity of Al



Phase field model development

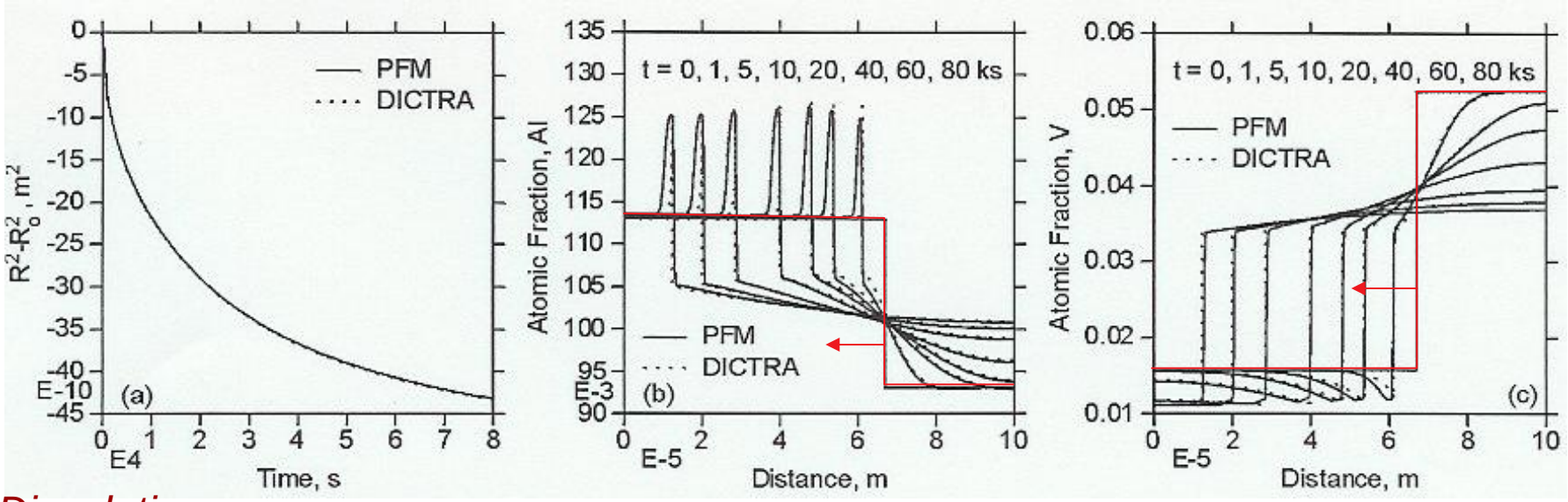
Phase Field Model

Constitutive equation construction

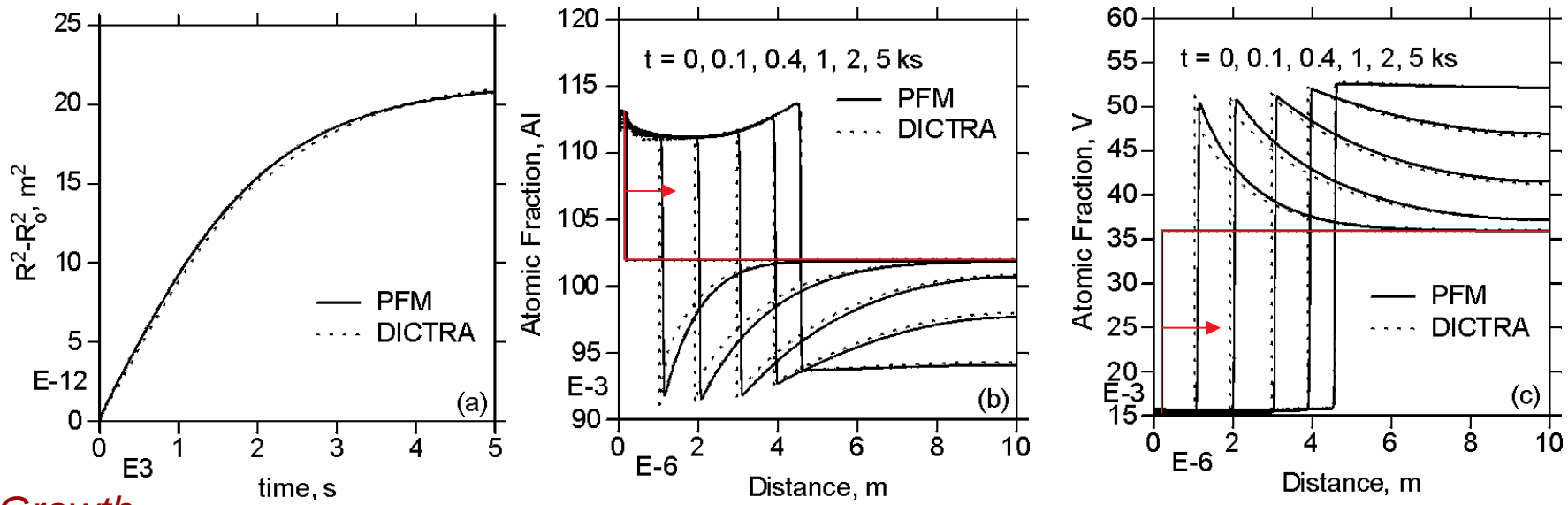
Scripta mater. 50(2004)471-476; *ibid*,50(2004)1145-1150

Quantitative comparison with DICTRA

Q. Chen et. al. ***Scripta mater.*** 50 (2004)471-476



Dissolution



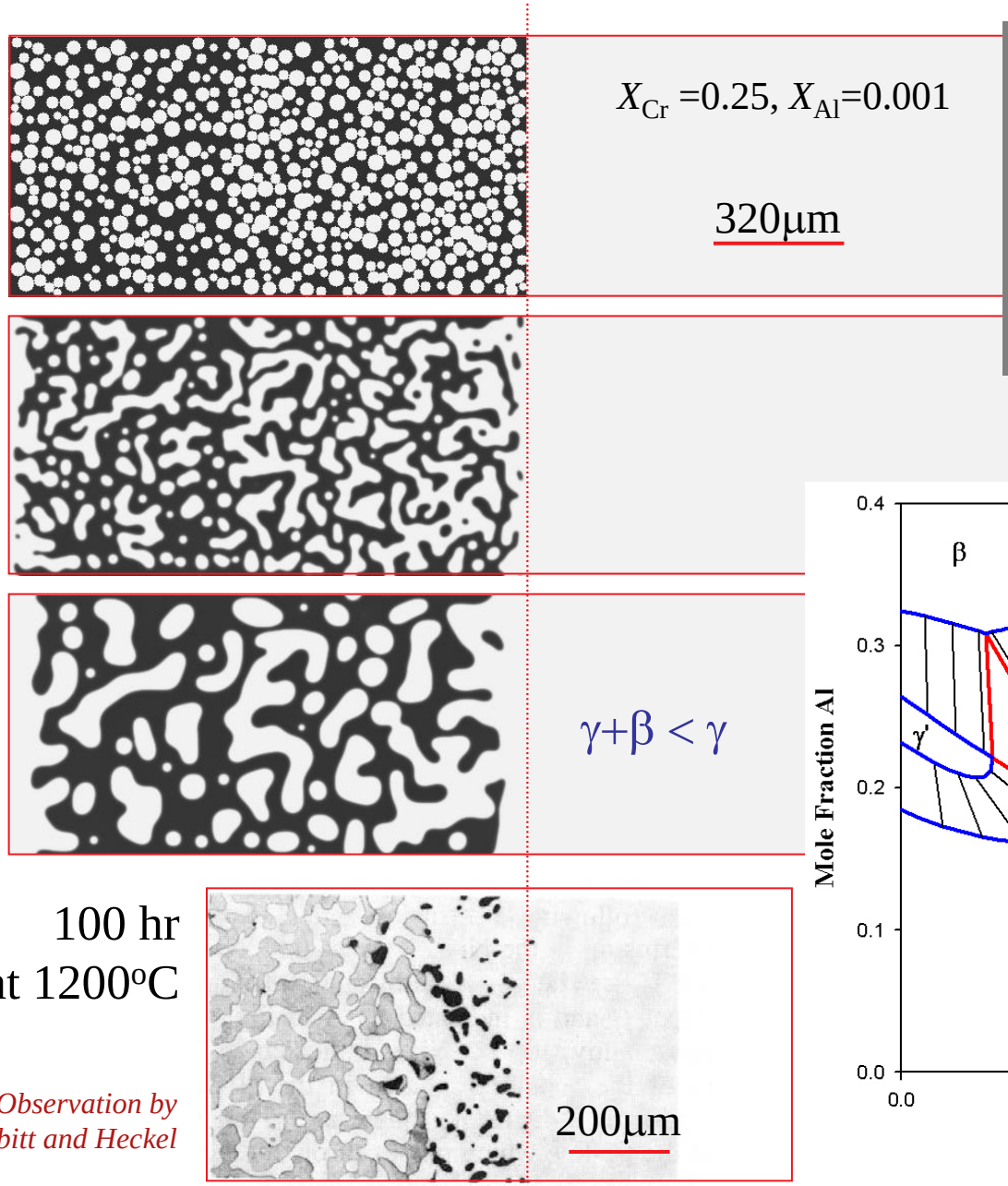
Growth

Interdiffusion Microstructure and Diffusion Path

0 hr

4 hr

25 hr

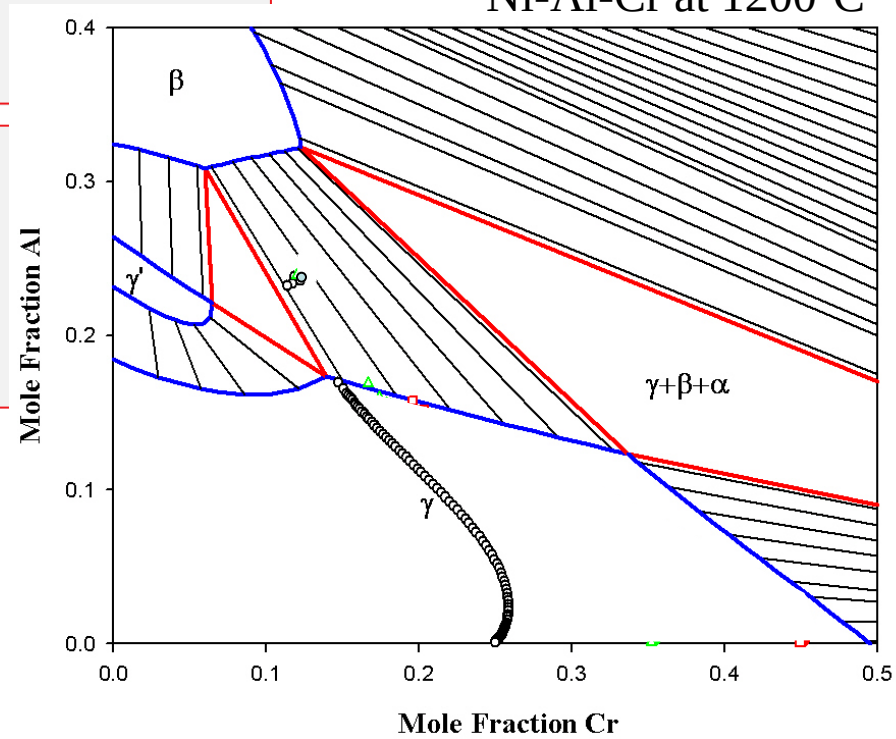


100 hr
at 1200°C

Exp. Observation by Nesbitt and Heckel

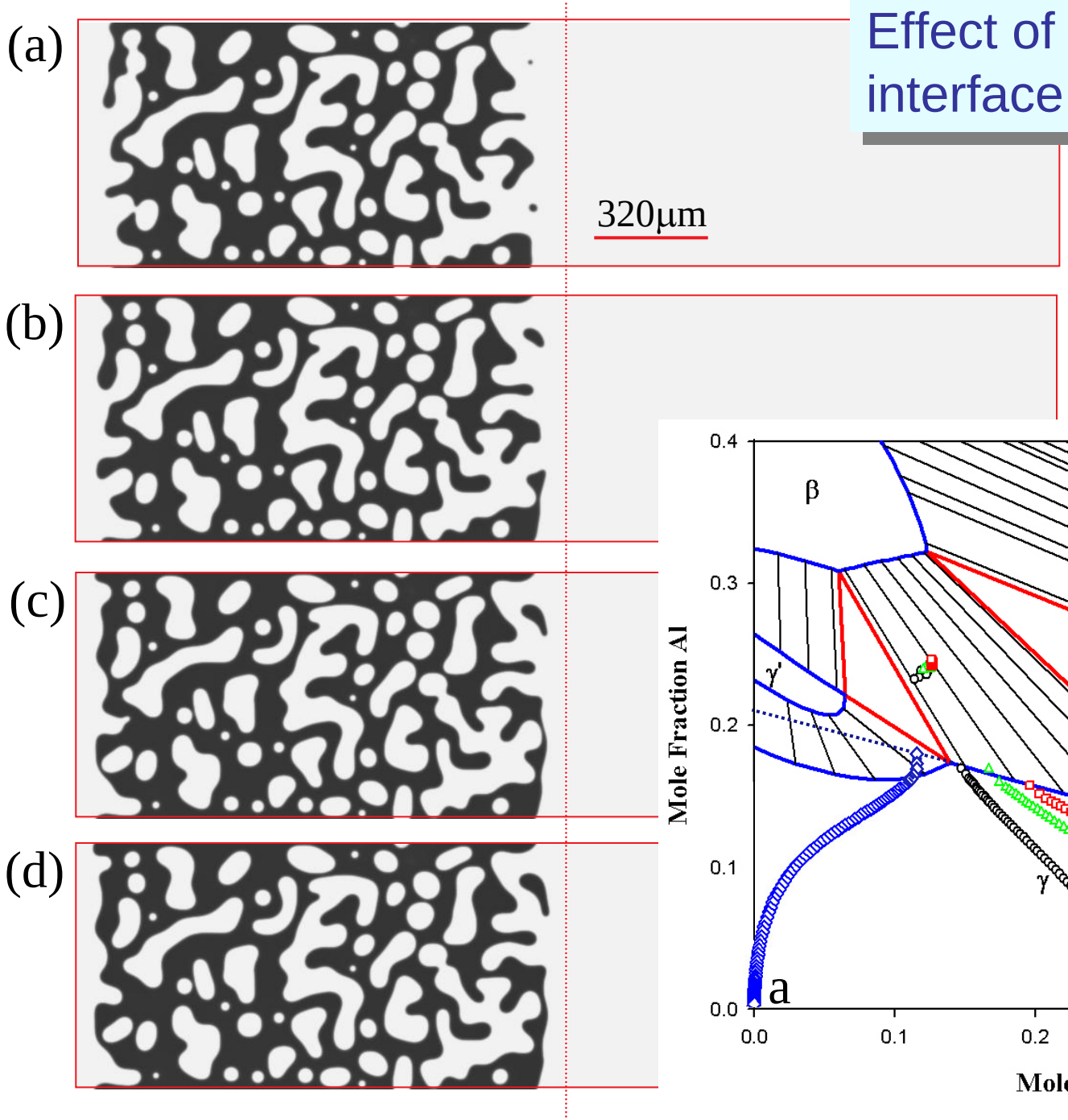
- Free energy data from Huang and Chang
- Mobilities in γ from A.EngstrÖm and J.Ågren
- Diffusivities in β from Hopfe, Son, Morral and Roming

Ni-Al-Cr at 1200°C

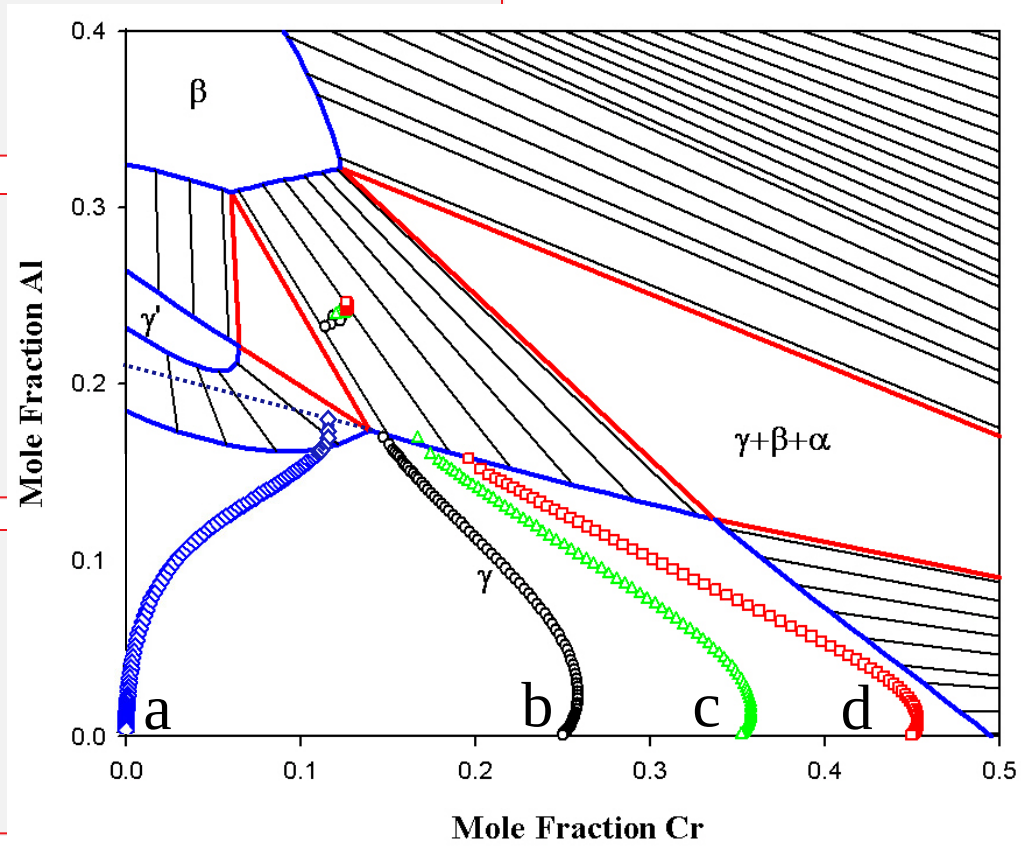


Effect of Cr content on interface migration $\gamma+\beta < \gamma$

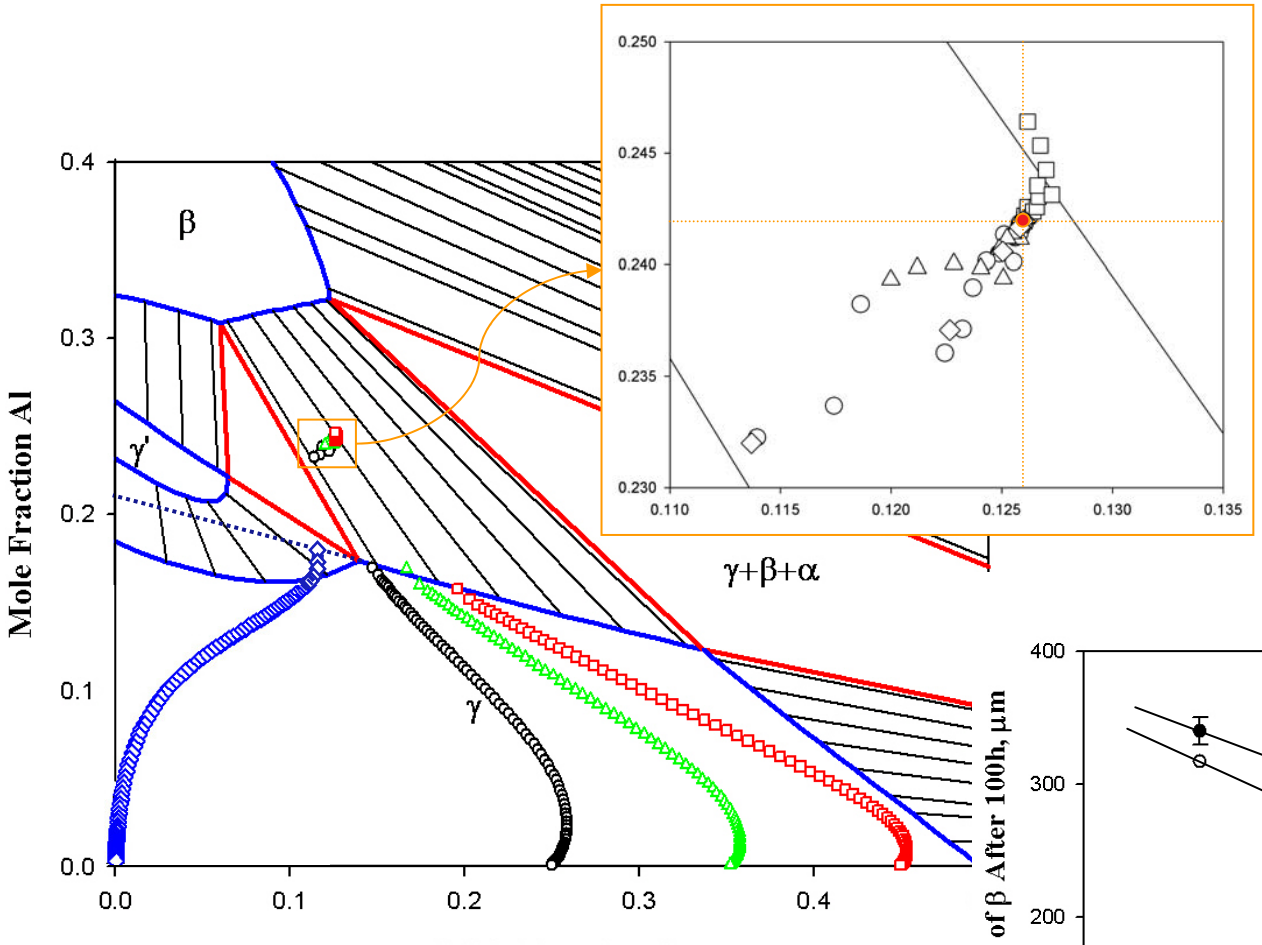
Annealing time:
25 hours

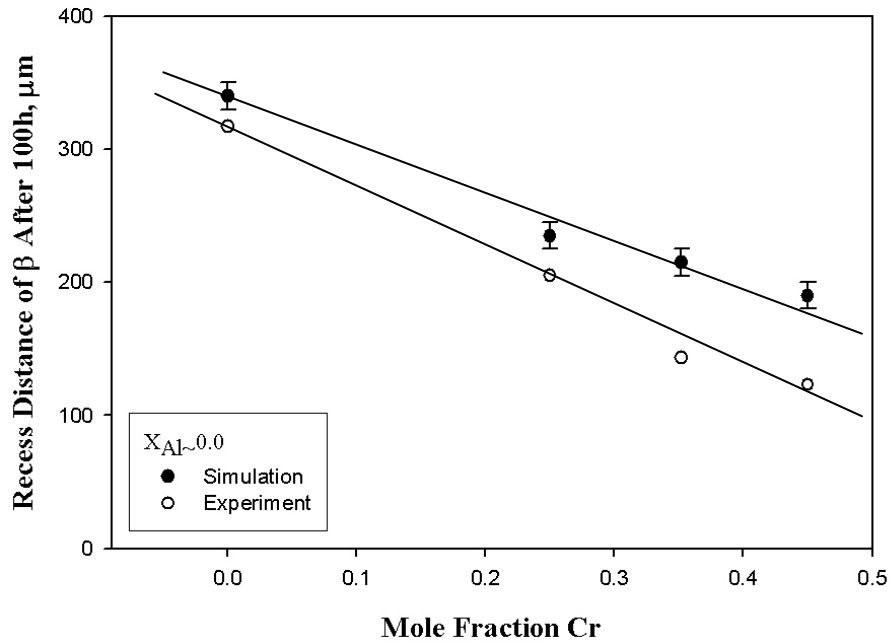


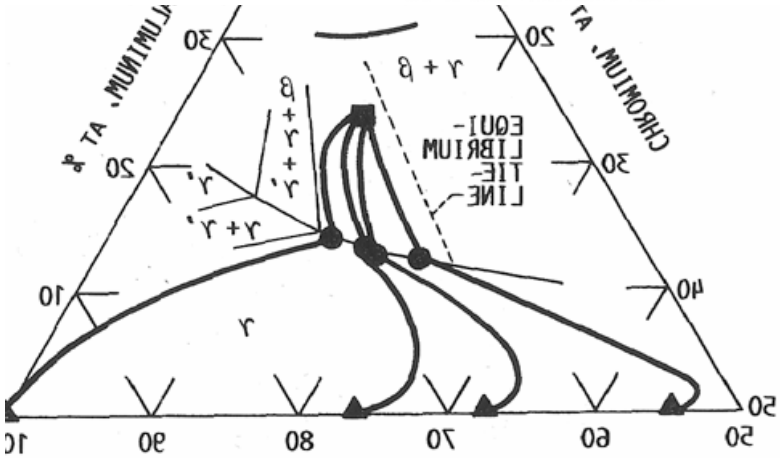
Ni-Al-Cr at 1200°C



Diffusion path and recess rate - comparison with experiment

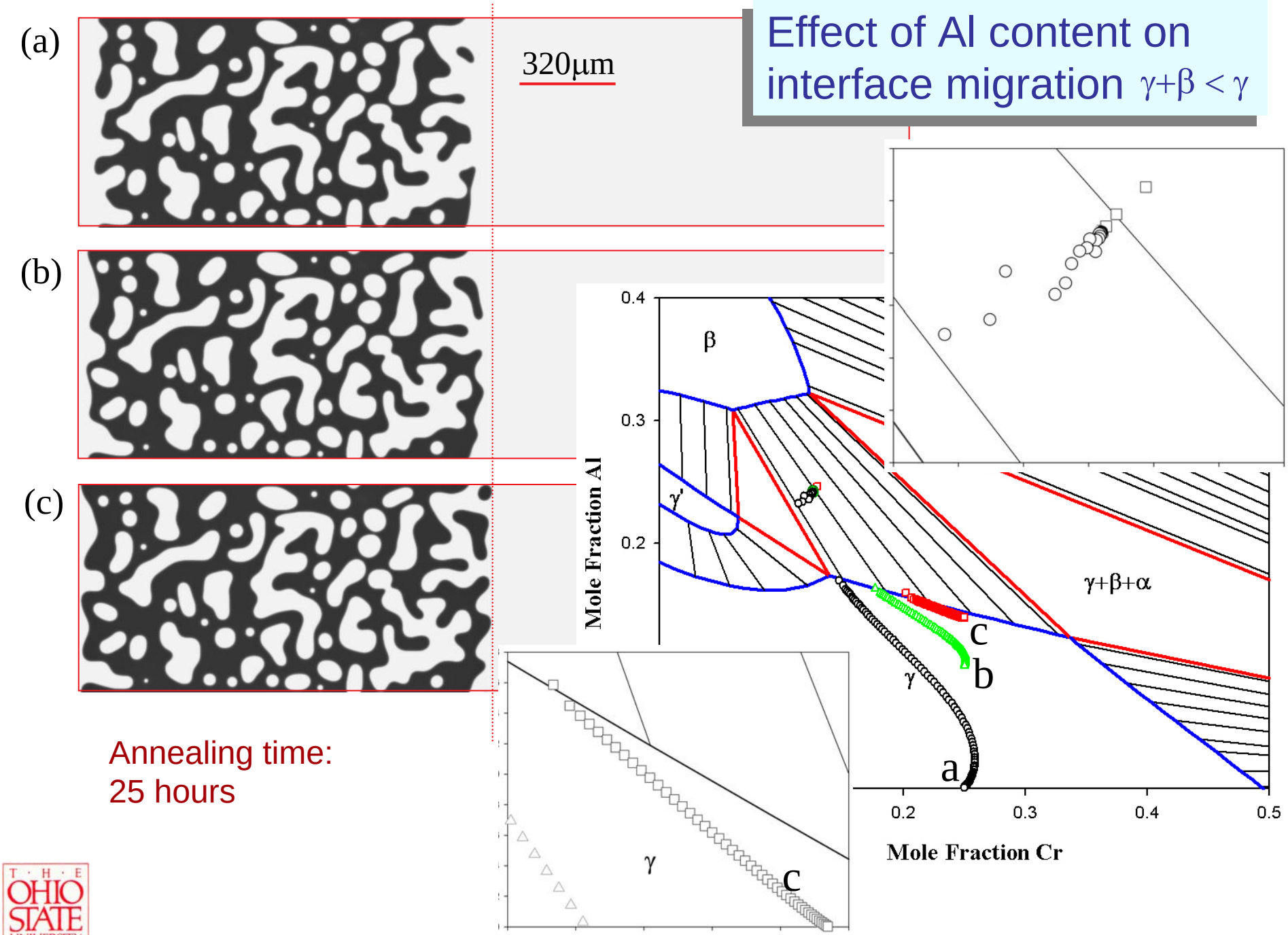






Exp. measurement by Nesbitt and Heckel

Effect of Al content on interface migration $\gamma+\beta < \gamma$



Annealing time:
25 hours

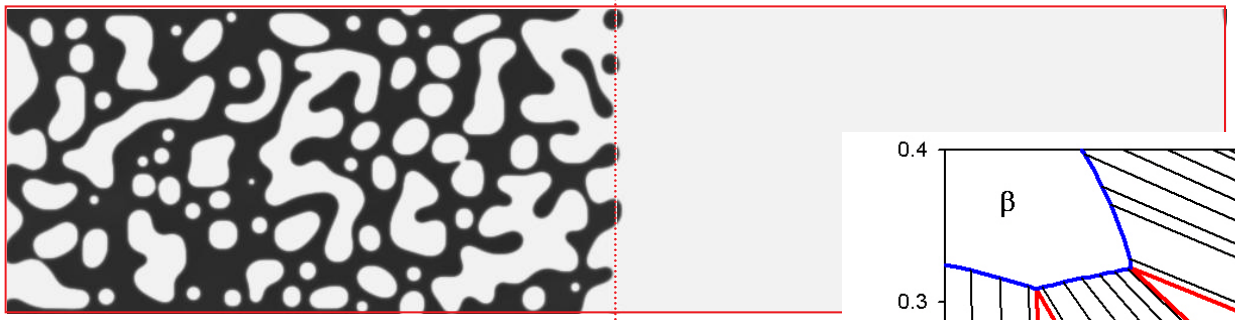
Effect of Al content on interface migration

Annealing time:
25 hours

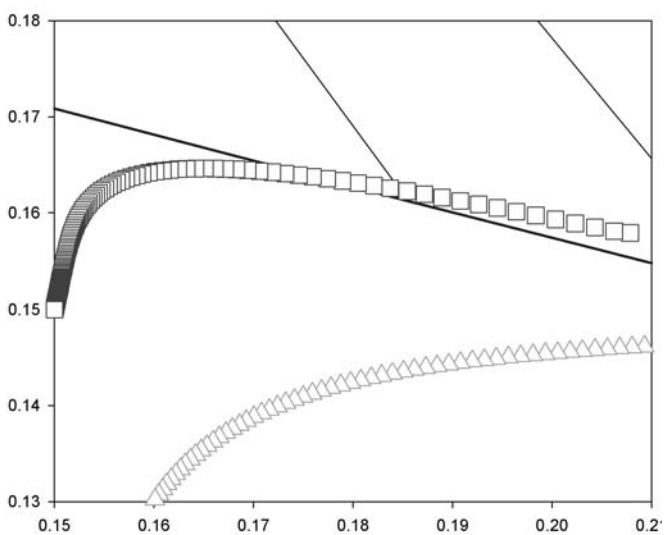
(a)

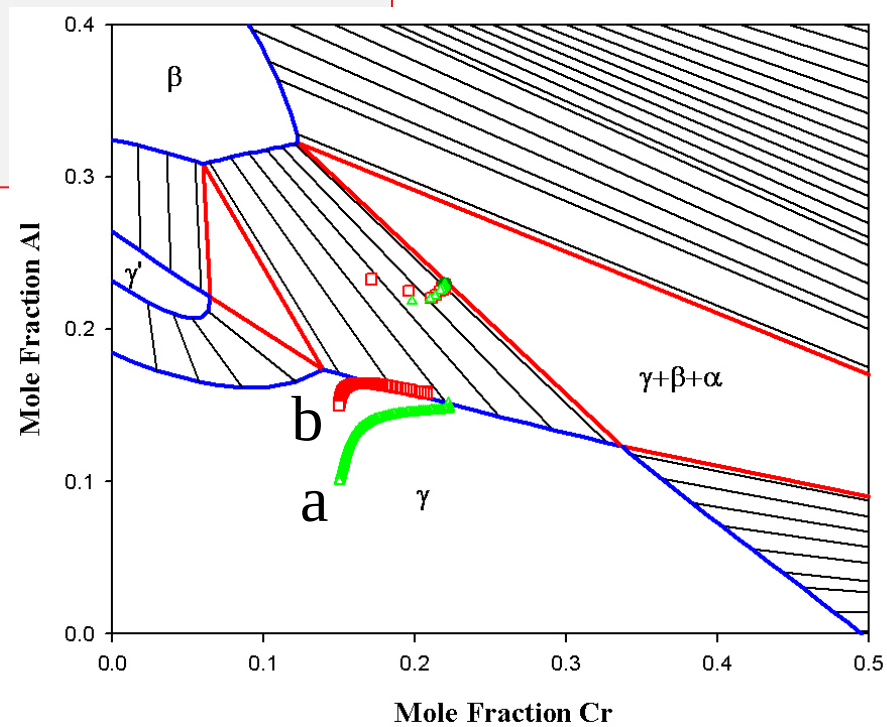
$\gamma+\beta < \gamma$

(b)

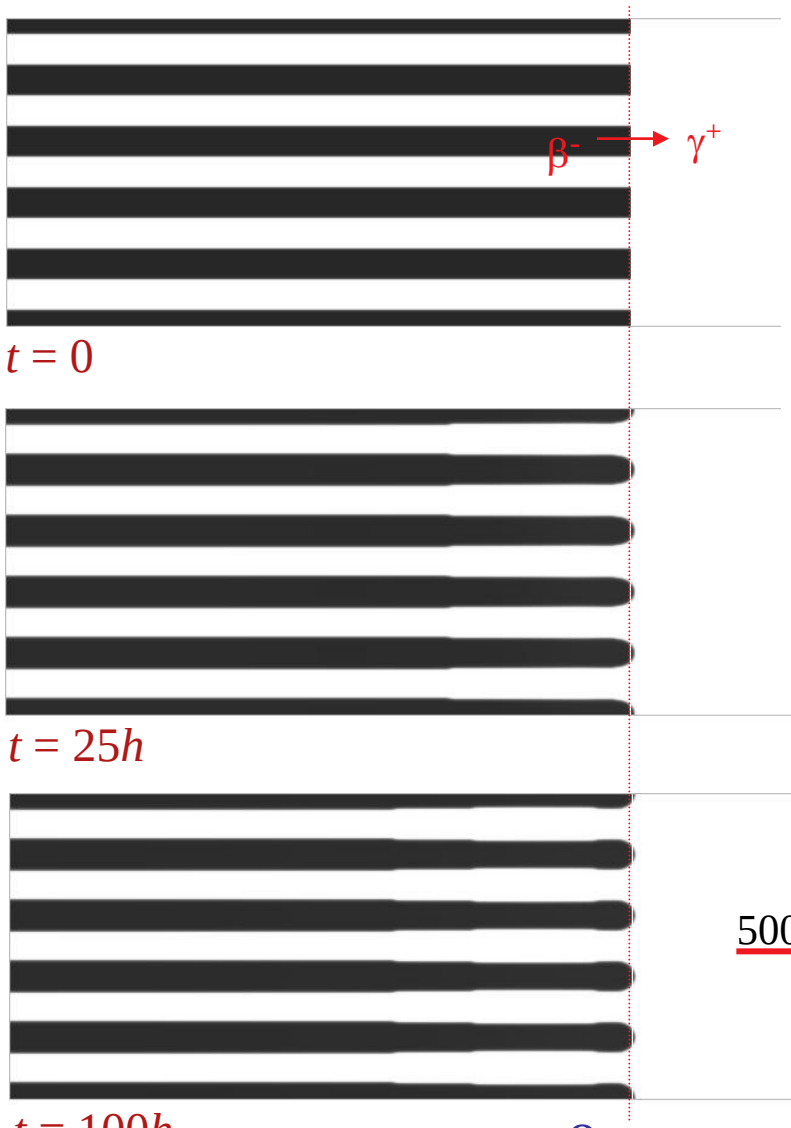


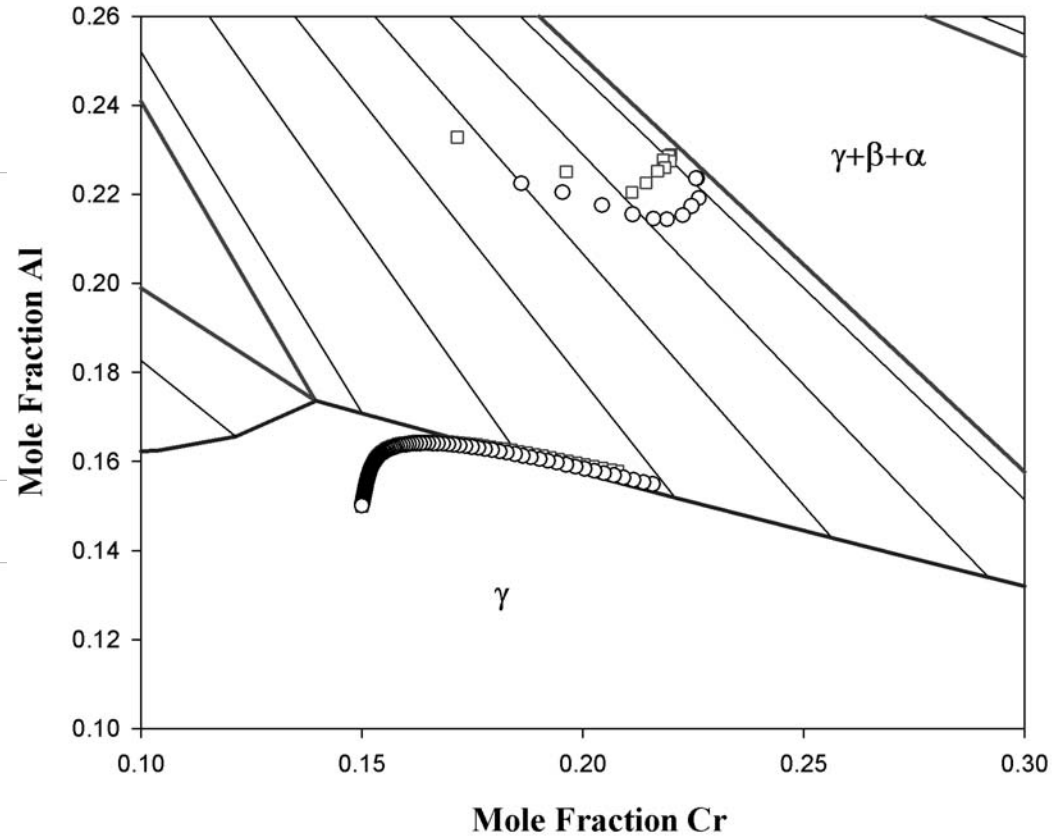
$\gamma+\beta > \gamma$



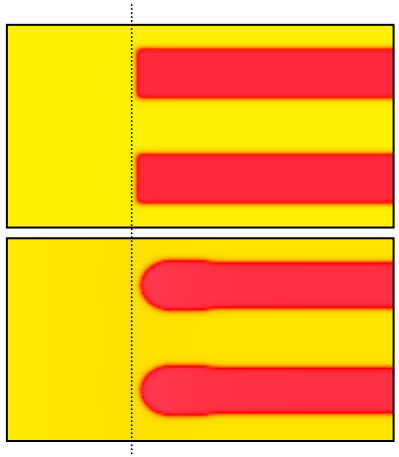


Shape of the Diffusion Path



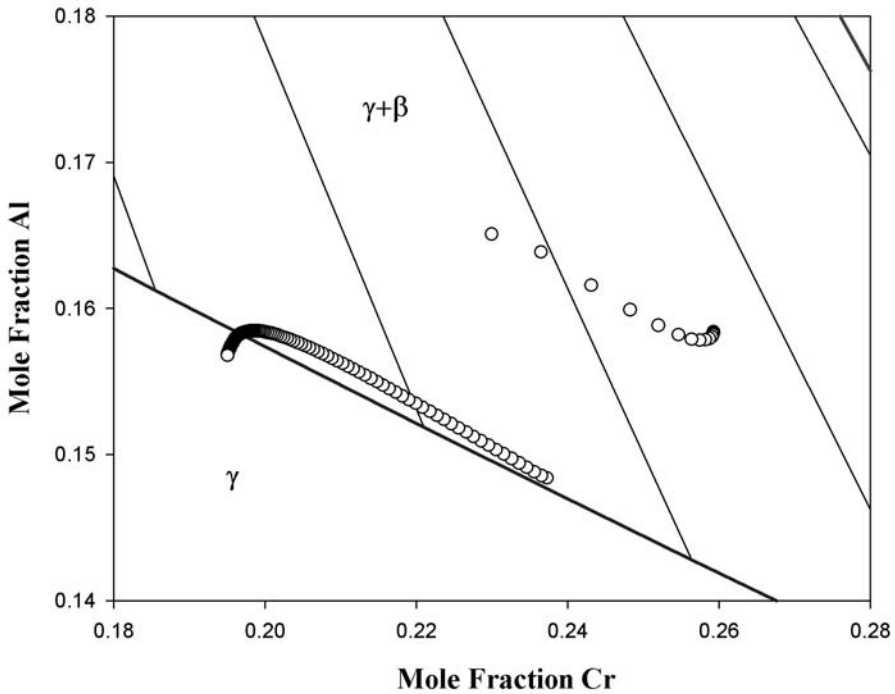


pure coarsening

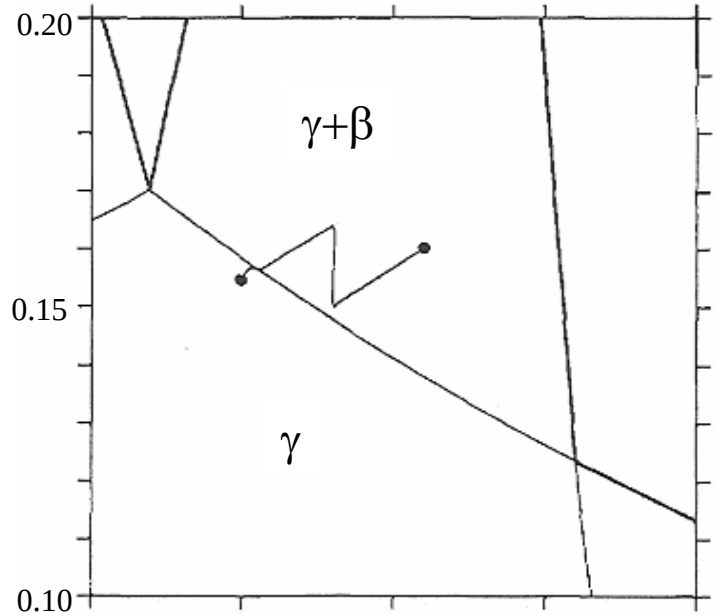


Shape of Diffusion Path - Comparison with DICTRA

$\gamma+\beta < \gamma$



$\gamma+\beta > \gamma$

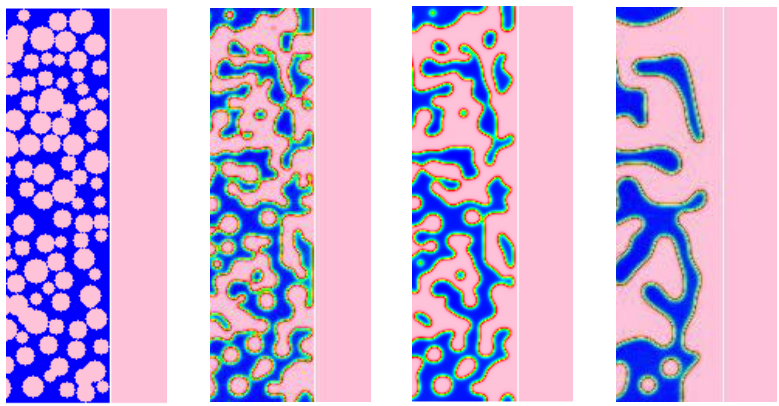


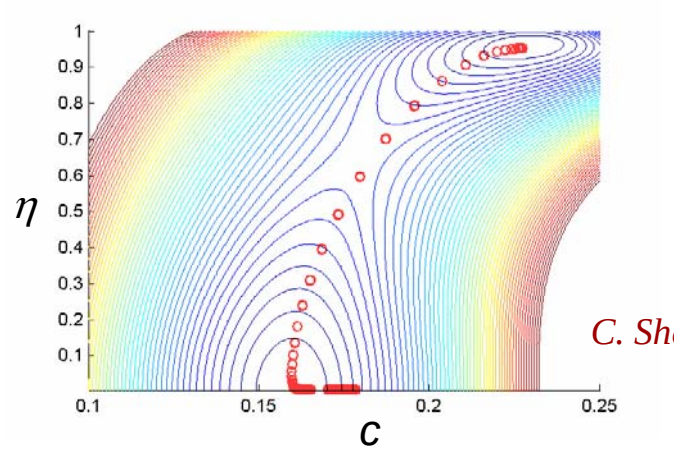
A.EngstrÖm, J. E. Morral and J.Ågren
Acta mater. 1997

Growth vs. Nucleation

Summary – Remaining Challenges

- Incorporation of nucleation
- Breaking the intrinsic length scale limit of quantitative phase field modeling
  - effect of surface energy, e.g., coarsening and coalescence





C. Shen et al., Scripta mater. (2004) **50**:1023-1028; *ibid*, 1029-1034.

- Quantitative comparison with experiment
  - accuracy of thermodynamic and mobility databases
  - accurate determination of average composition of multiphase microstructure in both simulation and experiment
  - Accurate determination of boundary position