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Phase-Field Modeling and Experimentation of Constituents Redistribution in Metallic Alloys

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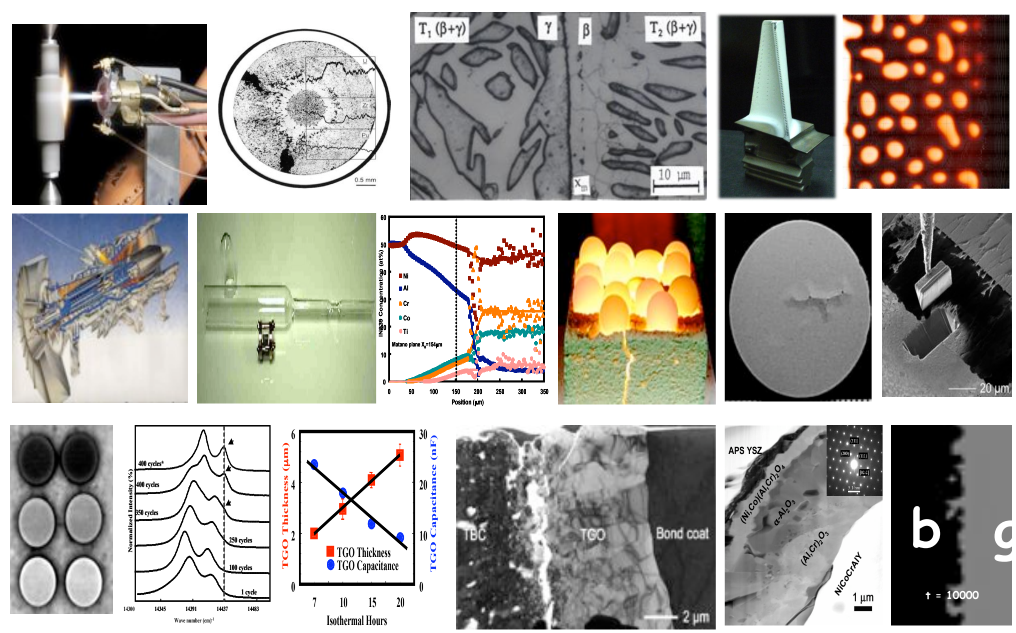


The Project Team

Dr. Rashmi Mohanty
Ph.D., UCF, 2009
Arcelor-Mittal Chicago

Senior Design Team for Building Thermotransport Apparatus. Billy, Sarah and Josh have committed to pursue M.S. in MSE.

Jayanta Kapat, Lockheed-Martin Professor of Mechanical Engineering with Heat Transfer Expertise: Heat transfer analysis for thermotransport apparatus.

Cheryl Xu, Assistant Professor of Mechanical Engineering with Controls Expertise: Computer-based active control of coolant for control of temperature and its gradient.

Objectives

- Development and implementation phase field approach to develop an applications-oriented, science-based microstructural model.

- Multicomponent and multiphase interdiffusion and thermotransport.

- Validation with selected experiments:

  - ✓ Thermotransport experimental apparatus
  - ✓ Determination and assessment of thermodynamic and kinetic parameters

Thermotransport or Thermomigration (Ludwig-Soret or Soret Effects)

- When a temperature gradient is applied to a homogeneous alloy of more than one component, a concentration gradient can develop and eventually reach a steady state, with the concentration gradient being a characteristic of the system.

- It can play an important role in microstructural stability, for example, in interconnects of electronic circuits, metallic nuclear fuel alloys, superalloys and coatings used in gas turbine engines, etc., where a significant temperature gradient is imposed, owing to increasing operating temperatures and/or reducing length scales of these systems and/or ingenuity in internal cooling methods.

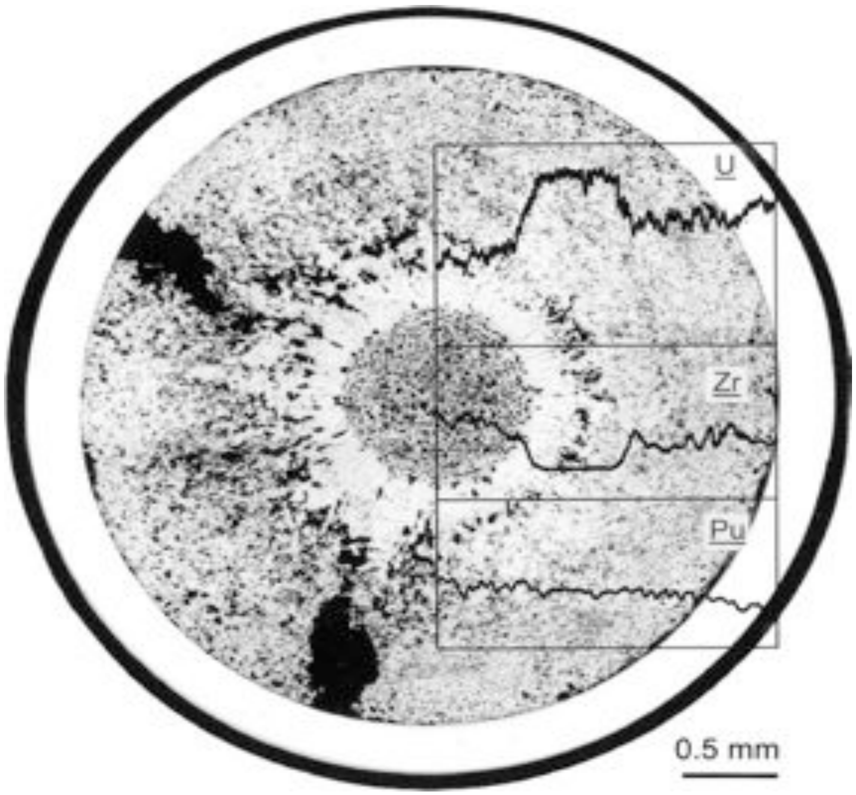
Thermotransport or Thermomigration (Ludwig-Soret or Soret Effects)

- Depending upon applications, temperature gradient imposed for a prolonged periods of time can vary from 20°C/mm up to 1500°C/mm although its magnitude can be reduced by means of various engineering solutions.

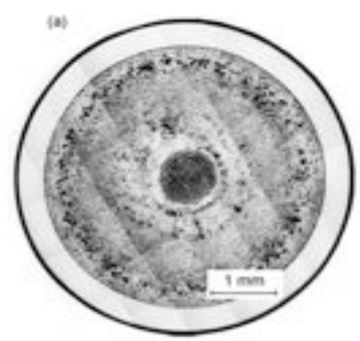
- There are two important parameters in the thermomigration study: the mobility of atoms present, i.e., diffusivity, and the heat of transport (Q_i^*), which is related to the amount of heat carried per atom of species *i*.

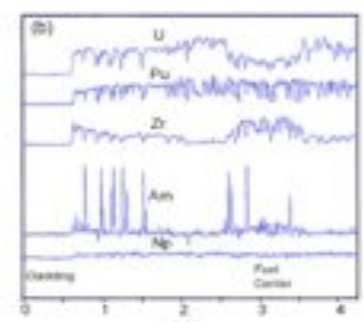
- The value of Q_i^* is the contribution of the flux of species *i* to the flux of heat, and determines the affinity of a species towards the cold or hot end.

Constituents Redistribution in Metallic Fuels



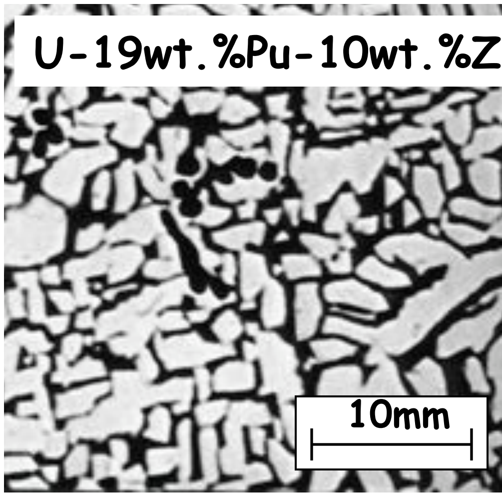
- Postirradiation observations typically show concentric microstructure:
  - ✓ Zr-enriched central zone.
  - ✓ Zr-depleted and U-enriched intermediate zone.
  - ✓ Slightly Zr-enriched outer periphery.





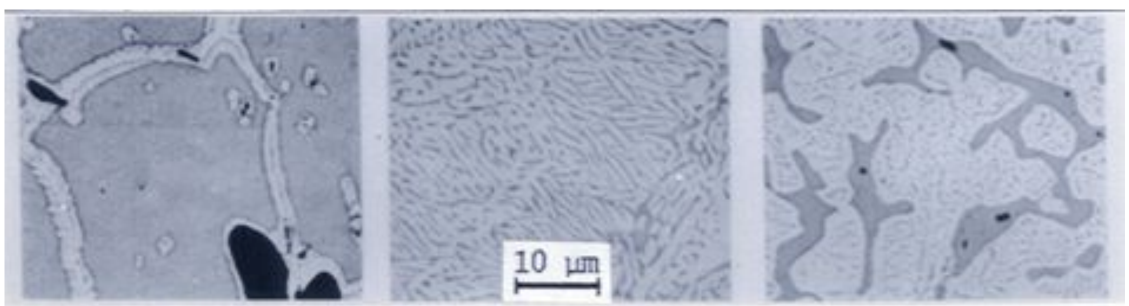
M.K. Meyer, S.L. Hayes, D.C. Crawford, R.G. Pahl, H. Tsai, Procd. ANS, 2001.
Y.S. Kim, G.L. Hofman, S.L. Hayes, Y.H. Sohn, JNM, 327 (2004) 27.

Experimental Investigation of Nonisothermal Thermotransport in U-Pu-Zr Alloy*



- δ (hexagonal) and ζ (tetragonal) phases at room temperature
- γ (bcc) phase at high temperature (T>650°C)
- Annealed under a temperature gradient of 220°C/cm for 41 days. (Carried out at Argonne National Lab.)
- Analysis by ND, SEM, EDS, EMPA

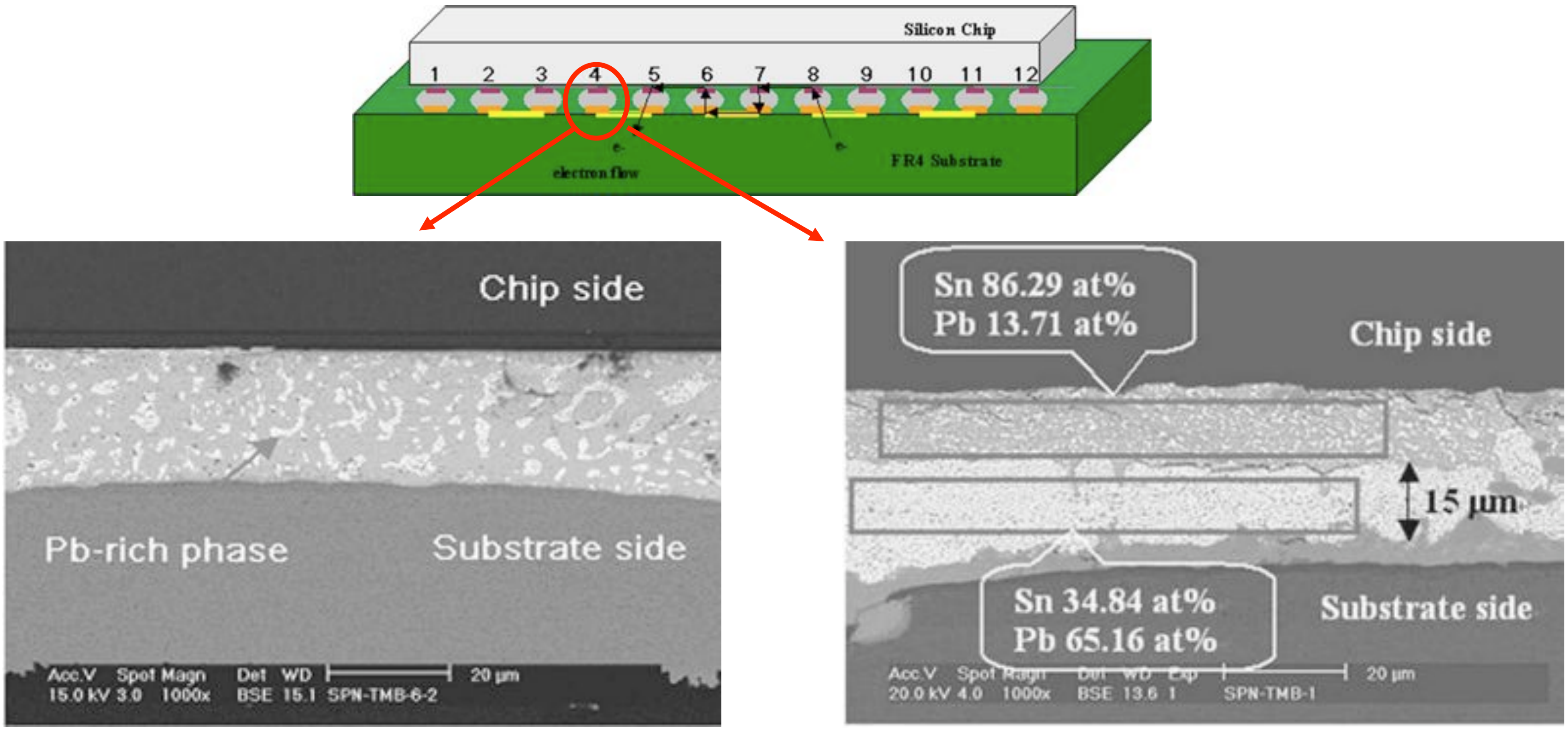
Near Hot Surface (T_{hot}=740°C)



Near Cold Surface (T_{cold}=600°C)

* Y. H. Sohn, M.A. Dayananda, G.L. Hofman, R.V. Strain, S.L. Hayes, JNM, 279 (2000) 317.

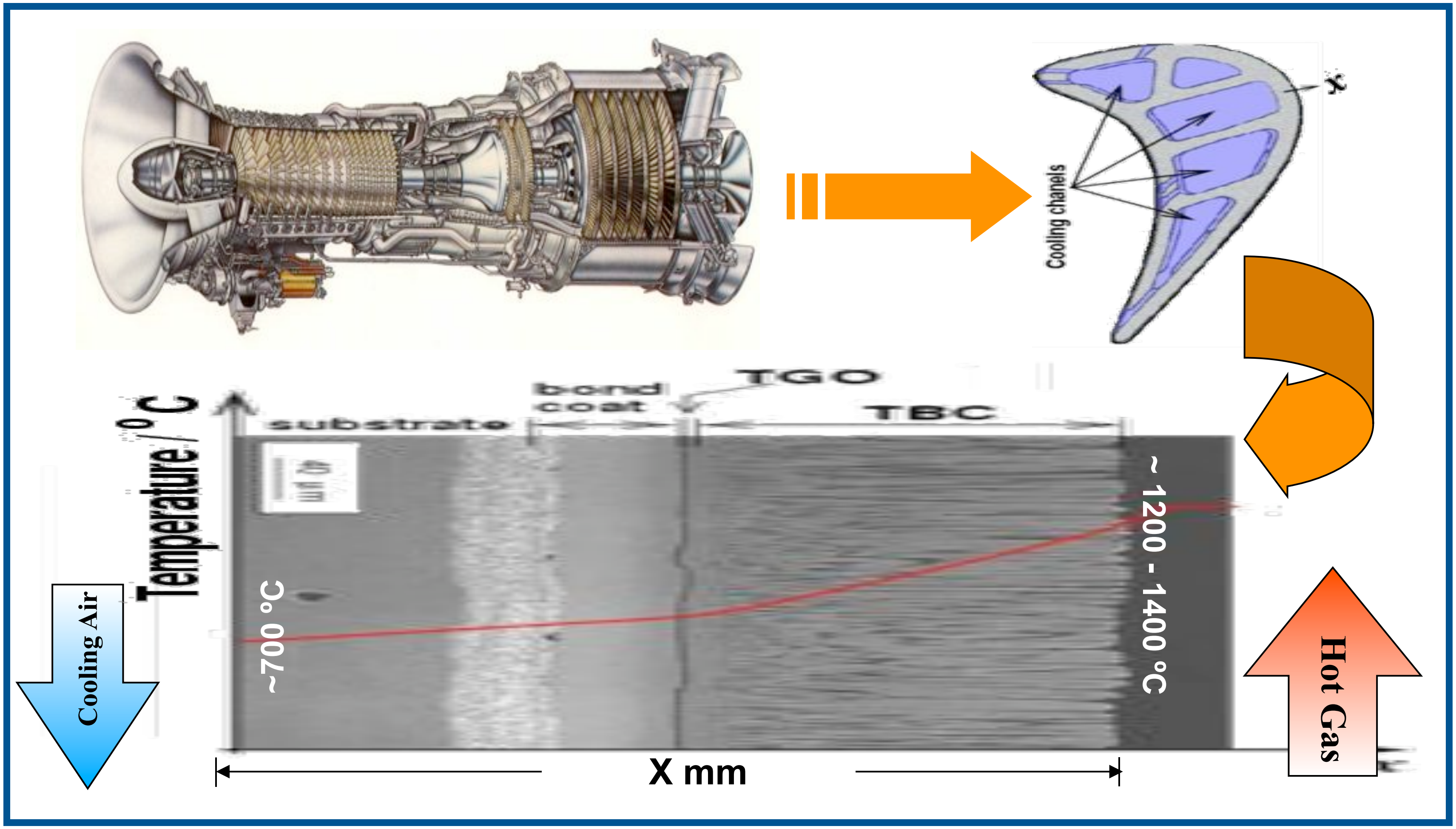
Constituent Redistribution in Interconnects



SEM micrographs of (a) original solder joint in between the chip and the substrate, (b) thermomigration (e.g., no electromigration) effect after 50 hours.

D. Yang, B. Y. Wu, Y. C. Chan, J. App. Phys., 102, 043502 (2007)

Constituent Redistribution in Turbine Blades (?)



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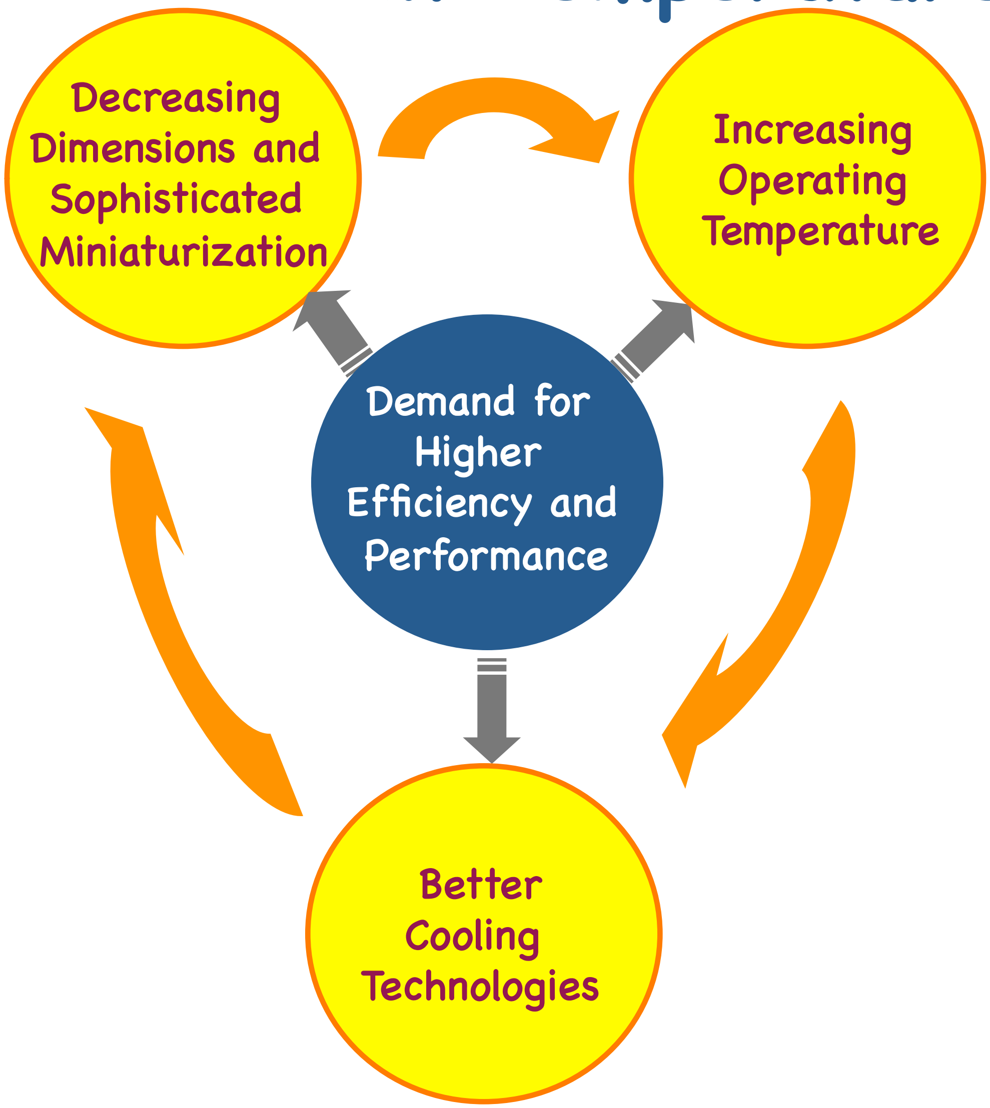
Constituent Redistribution in Temperature Gradient



- Decreasing Dimensions / Sophisticated Miniaturization of Engineered Components.
- Increasing Operating Temperatures (e.g., Efficiency).
- Aggressive and Advanced Cooling Technologies.

Phase Field Approach (Presented at '08 NIST Workshop)

$$J_k = \sum_{i=1}^{n-1} L_{ki}\left(X_i - X_n + Q_i^* X_q\right)$$

Onsager's Formalism of fluxes under gradients of chemical potential and temperature.

$$\tilde{J}_A = -\rho c(1-c)\left[c\beta_A + (1-c)\beta_B\right]\nabla\mu_A^{eff} + \rho c(1-c)\left[\beta_A Q_A^{*'} - \beta_B Q_B^{*'}\right]X_q$$
$$= -M_c \nabla\mu_A^{eff} + M_Q X_q$$

On Laboratory Fixed Reference with Gibbs-Duhem Relations.

$$\tilde{J}_B = -\tilde{J}_A = -M_c \nabla\mu_B^{eff} + M_Q \frac{\nabla T}{T}$$

$$\frac{\partial n_B}{\partial t} = \nabla \cdot \left[V_m M_c \nabla\left(\frac{\partial f}{\partial c} - 2\kappa_c \nabla^2 c\right) - M_Q \frac{\nabla T}{T}\right]$$

$$M_c = \rho c(1-c)\left[c\beta_A + (1-c)\beta_B\right]$$

$$M_Q = \rho c(1-c)\left[\beta_A Q_A^{*'} - \beta_B Q_B^{*'}\right]$$

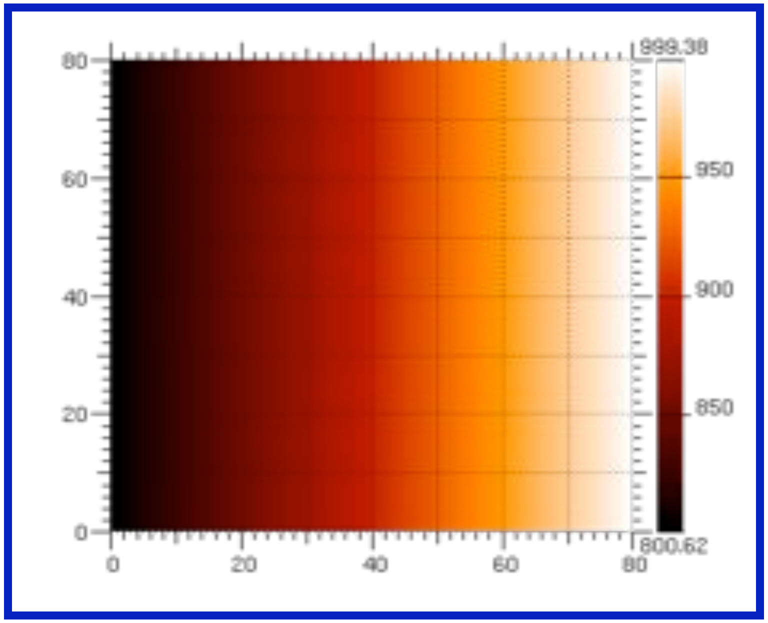
$$\beta_i = A_i \exp\left(\frac{-Q_i}{RT}\right)$$

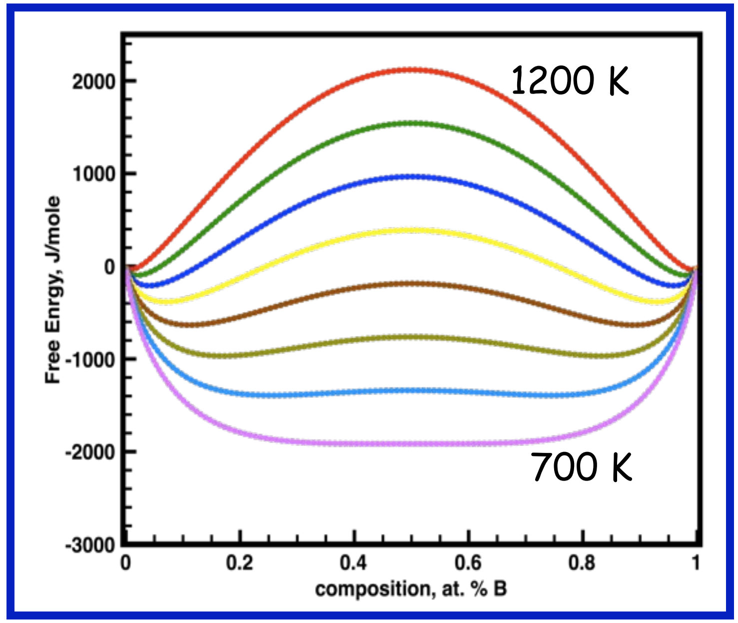
$$Q_i^* = B_i + C_i T$$

1. Howard R. E. and Lidiard A. B., "Matter Transport in Solids", Rep. Prog. Phys., 27, p.161

Temperature Distribution and Free Energy

- **The temperature field obeys Laplace's equation:** $\nabla^2 T = 0$
- **Boundary conditions:** $J_q \cdot \hat{n} = 0, T\big|_{x=0} = T_{\min}, T\big|_{x=L} = T_{\max}$





Applied temperature gradient and the free energy vs. composition curves at different temperatures.

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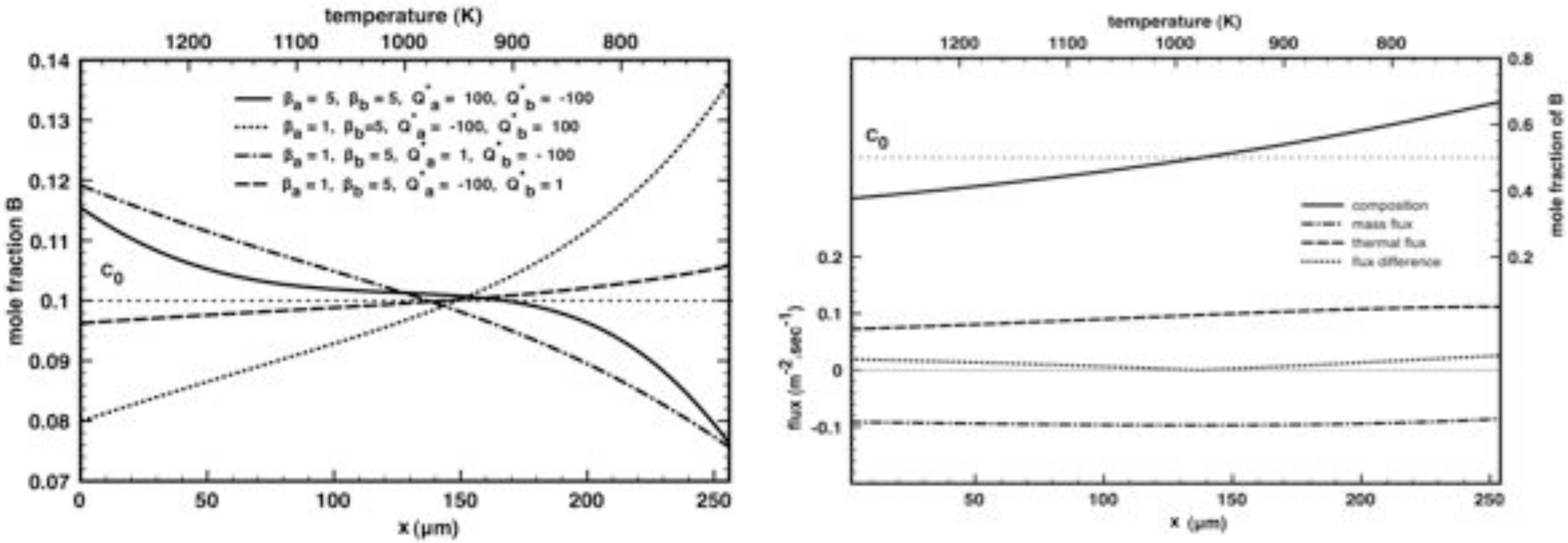
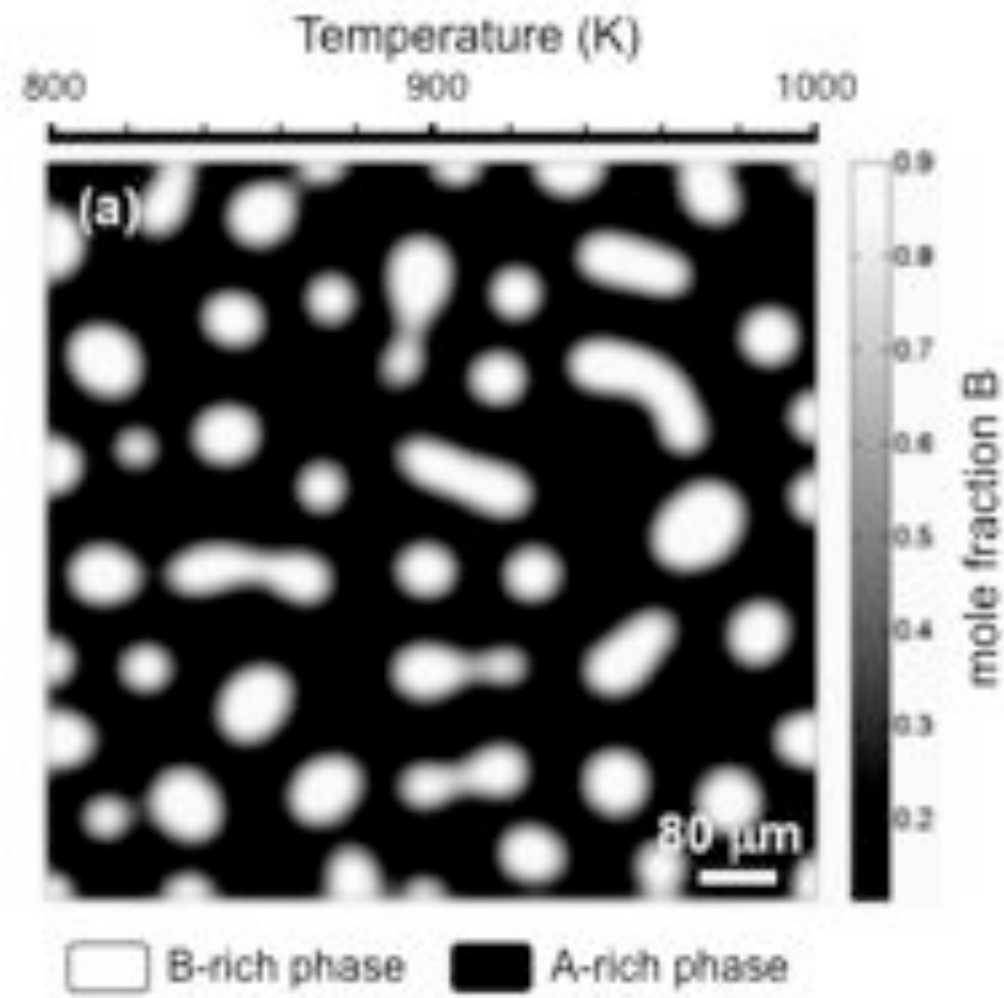
Phase Field Simulation of Thermotransport Substitutional Single-Phase



- Composition profiles of component B developed under temperature gradient for various combinations of β and Q^*.
- The concentration gradient depends on the initial composition and the values of atomic mobility as well as heat of transport.
- Steady state can be reached after a long time of anneal under thermal gradient.
- Similar magnitude but opposite direction of flux contributed by gradients of temperature and concentration.

Phase Field Simulation of Thermotransport Substitutional Two-Phase Alloy

NIST Collaboration for FIPY (Dr. J. Guyer)



Initial Microstructure from Ideal Solid Solution, A-B.

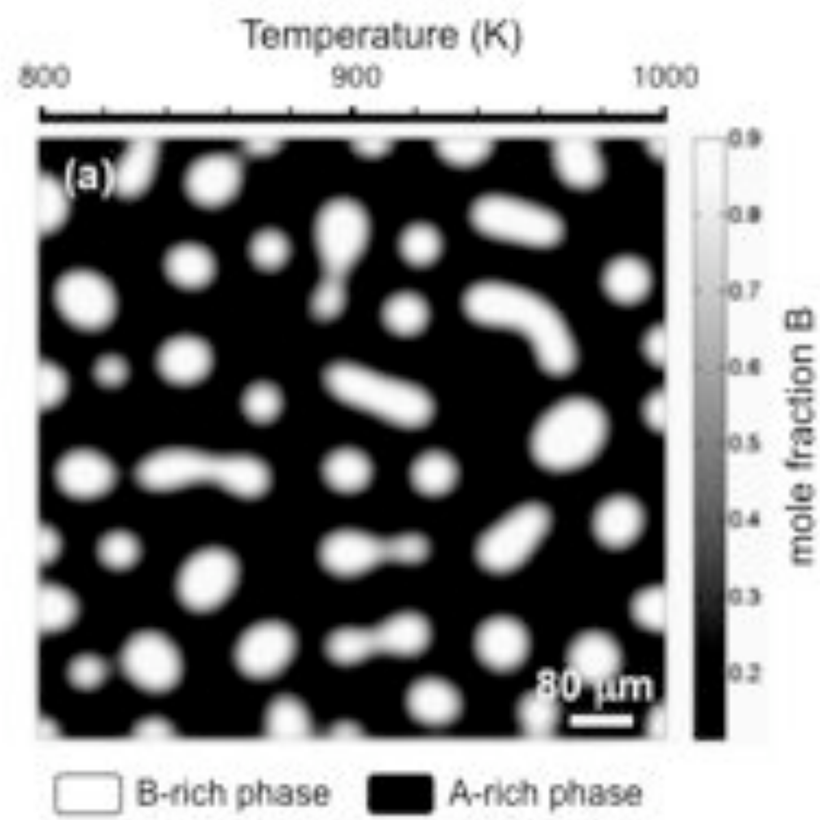
Bright Region: B-rich Phase
Dark Region: A-rich Phase

B-rich Nuclei Introduced and Annealed to Minimize Coarsening Effects.

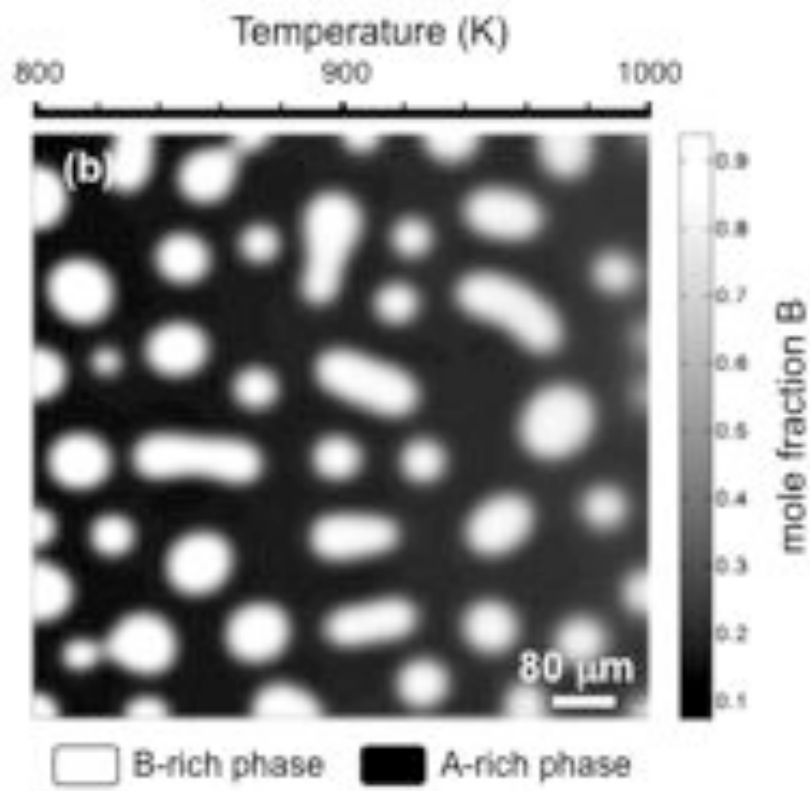
FIPY Partial Differential Equation (PDE) Solver using a Finite Volume Approach.

Phase Field Simulation of Thermotransport Substitutional Two-Phase Alloy

NIST Collaboration for FIPY (Dr. J. Guyer)

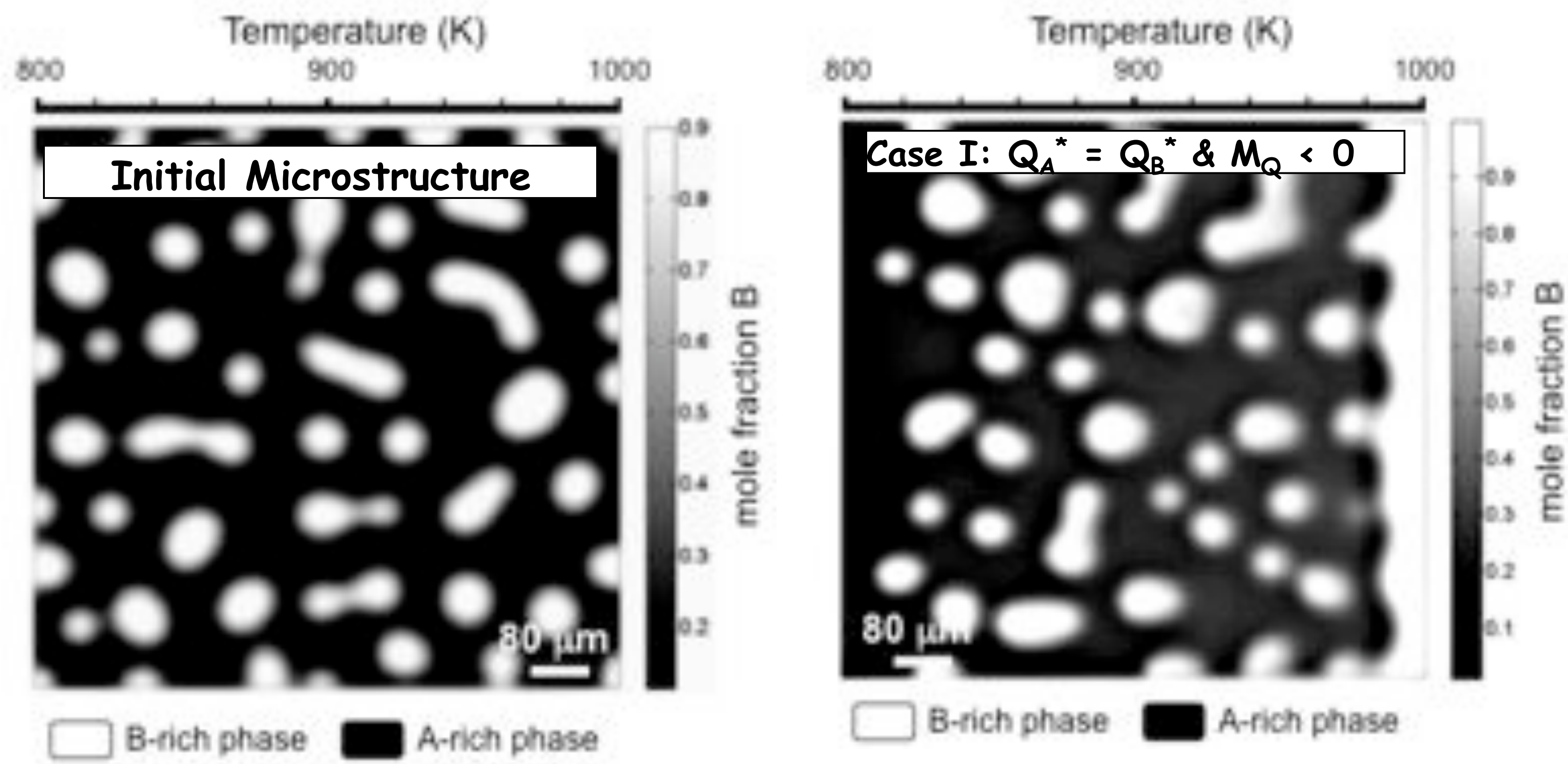


Initial Microstructural



Microstructural Evolution due to Temperature Difference (without Thermotransport Effect, M_Q = 0)

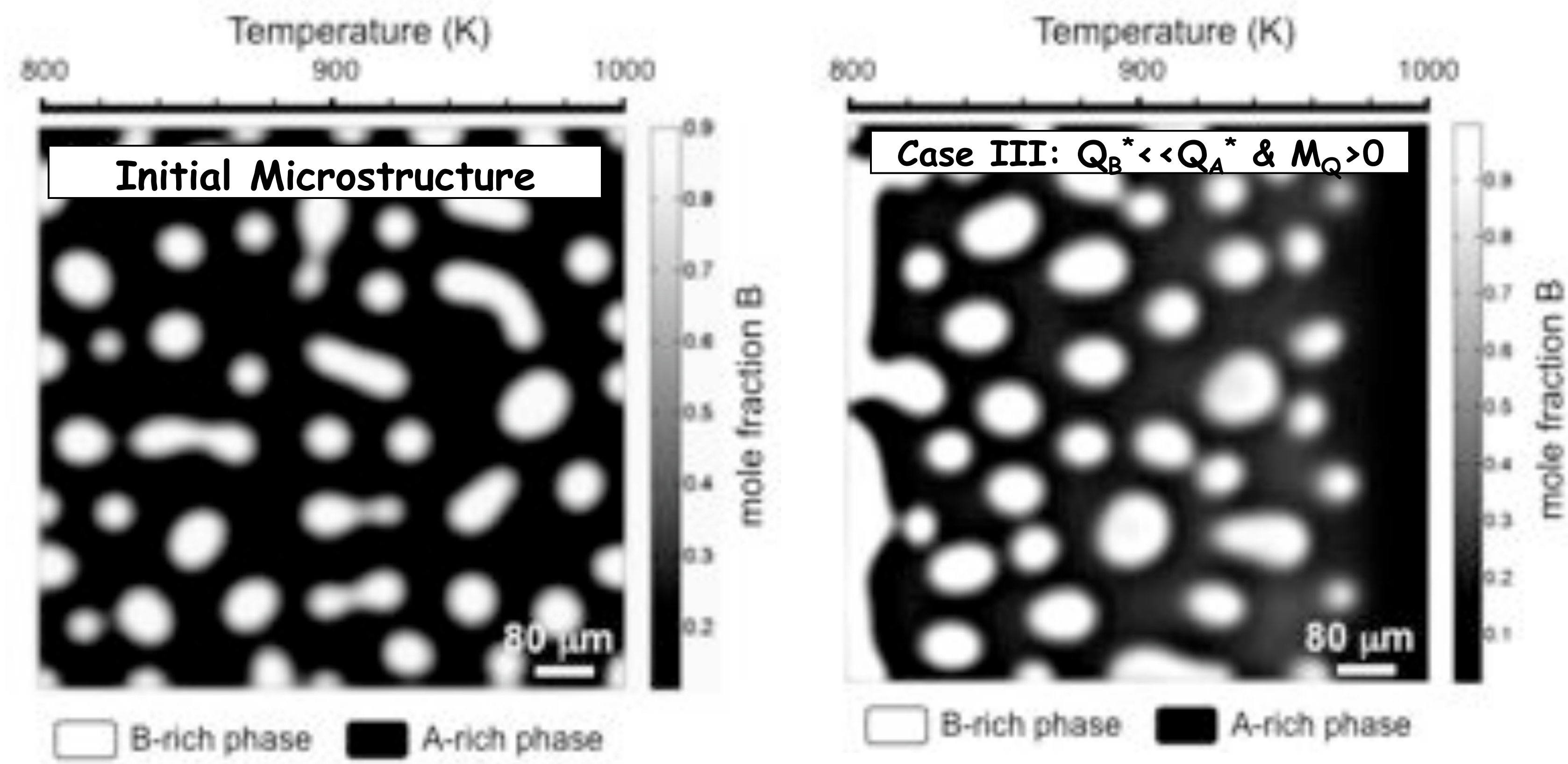
Phase Field Simulation of Thermotransport Substitutional Two-Phase Alloy
NIST Collaboration for FIPY (Dr. J. Guyer)



- Preferential movement of B atoms towards the hot end and A atoms towards the cold end.
- Phase redistribution occurs with B-rich and A-rich single phase regions forming at the hot and cold ends.

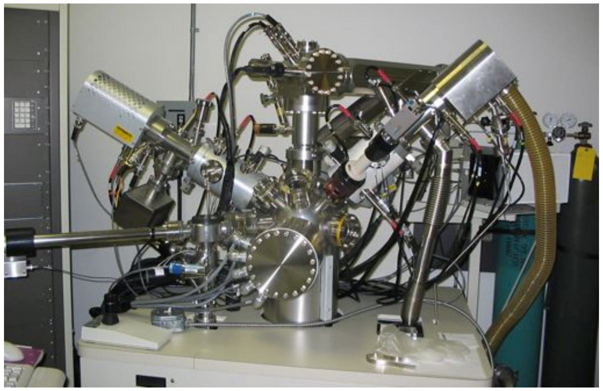
Phase Field Simulation of Thermotransport Substitutional Two-Phase Alloy

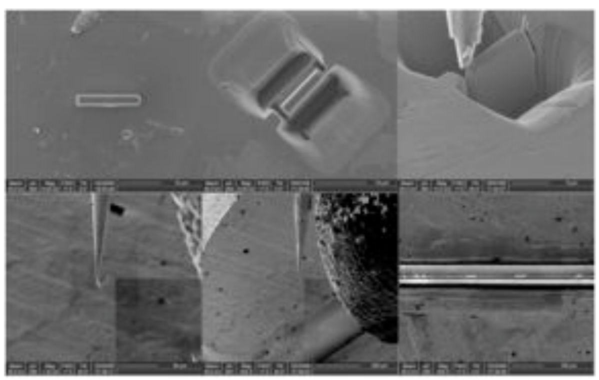
NIST Collaboration for FIPY (Dr. J. Guyer)



- Preferential movement of B atoms towards the cold end and A atoms towards the hot end.
- Phase redistribution occurs with B-rich and A-rich single phase regions forming at the hot and cold ends.

Experimental Diffusion Work at UCF

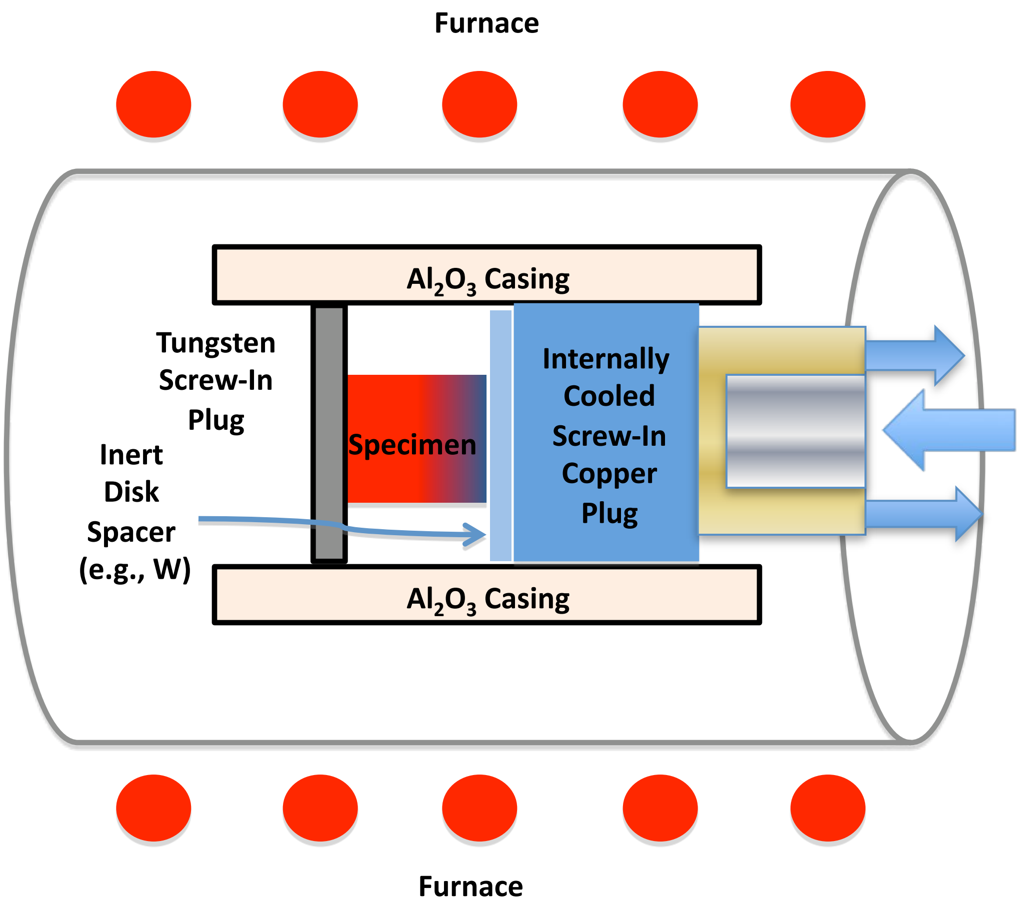




- Alloy Casting by Vacuum Induction Melting, Chill Casting and/or Tri-Arc Melting Furnace.
- Homogenization Heat Treatment.
- Microstructure, Phase Constituents and Compositional Analysis.
- Assembled with Kovar Steel Jigs.
- Encapsulate in Quartz Tube (Vacuum or Ar-Filled) After Several Vacuum-Hydrogen Flush.
- Diffusion Anneal Using Three-Zone Tube Furnace.
- Metallographic Preparation and Microstructural Analysis.
- Compositional Analysis by Electron Probe Microanalysis (EMPA).
- Depth Profiling by Secondary Ion Mass Spectroscopy (SIMS).
- Interfacial Analysis by Transmission Electron Microscopy (TEM) vis Focused Ion Beam (FIB) In-Situ Lift-Out (INLO)

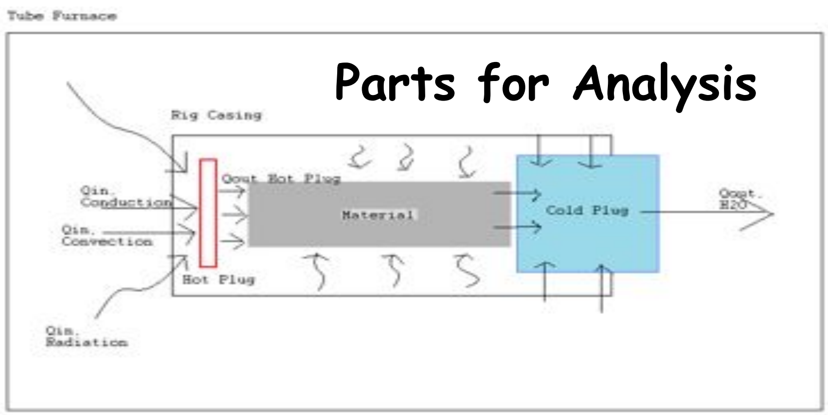
Thermotransport (aka Thermomigration, Soret effect)

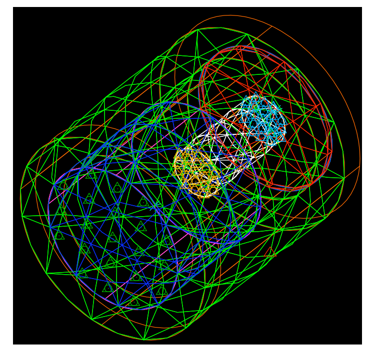
Experimental Set-Up



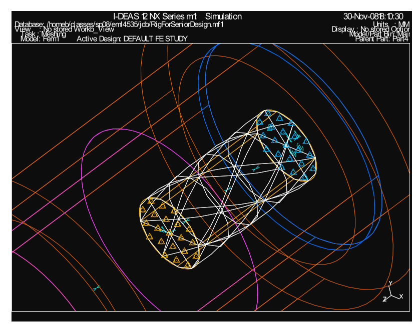
- Fits into Tube Furnace – Tube Diameter of 3 inches
- Materials Selection for Durability and Heat Conduction/Insulation.
- User-Friendly Computer Active-Control of Coolant Flow based on Temperature Measurement (and so Temperature Gradient Control).
- Numerical Heat Transfer Analysis for Temperature Distribution.

Energy Balance and Heat Transfer Analysis (Analytical and Numerical) of the Thermotransport Rig

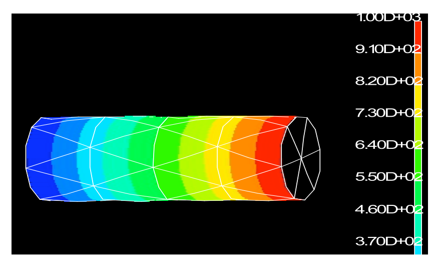




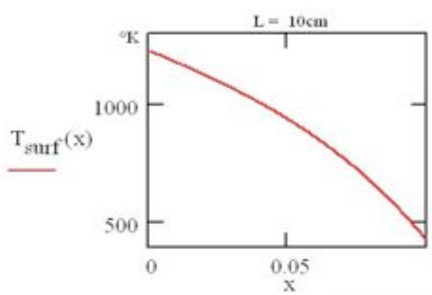
Geometry Constraints

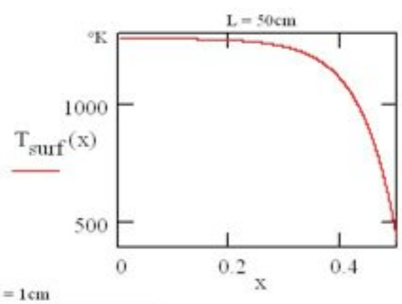


Material Constraints

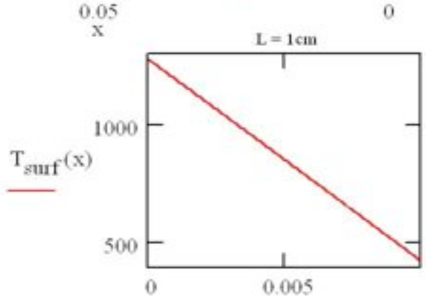


Temperature Distribution

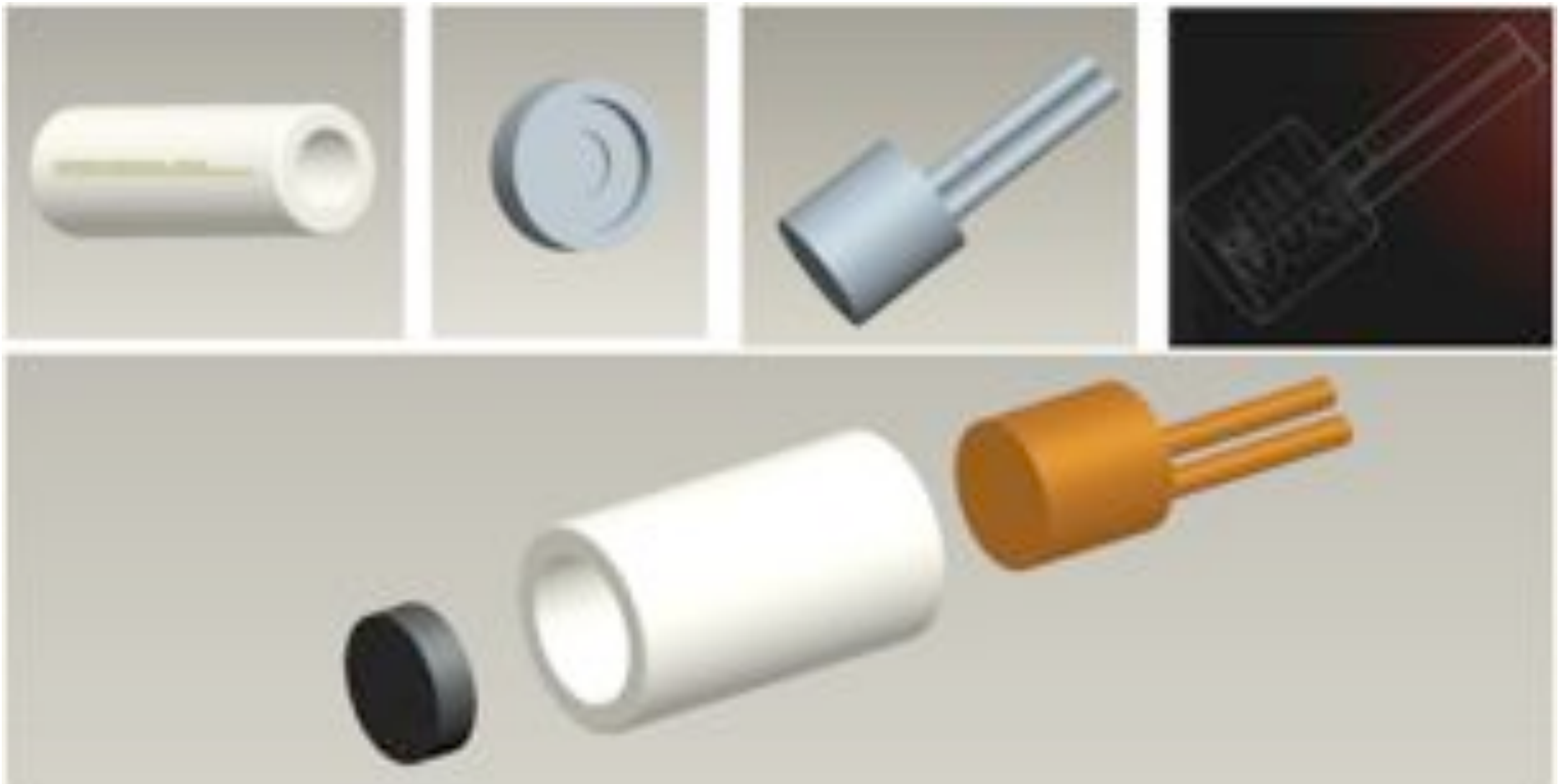


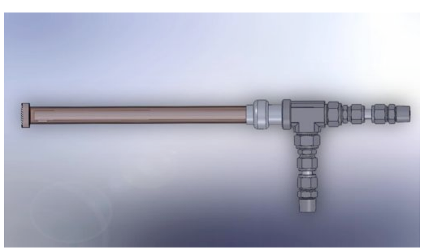


Analytical Solutions



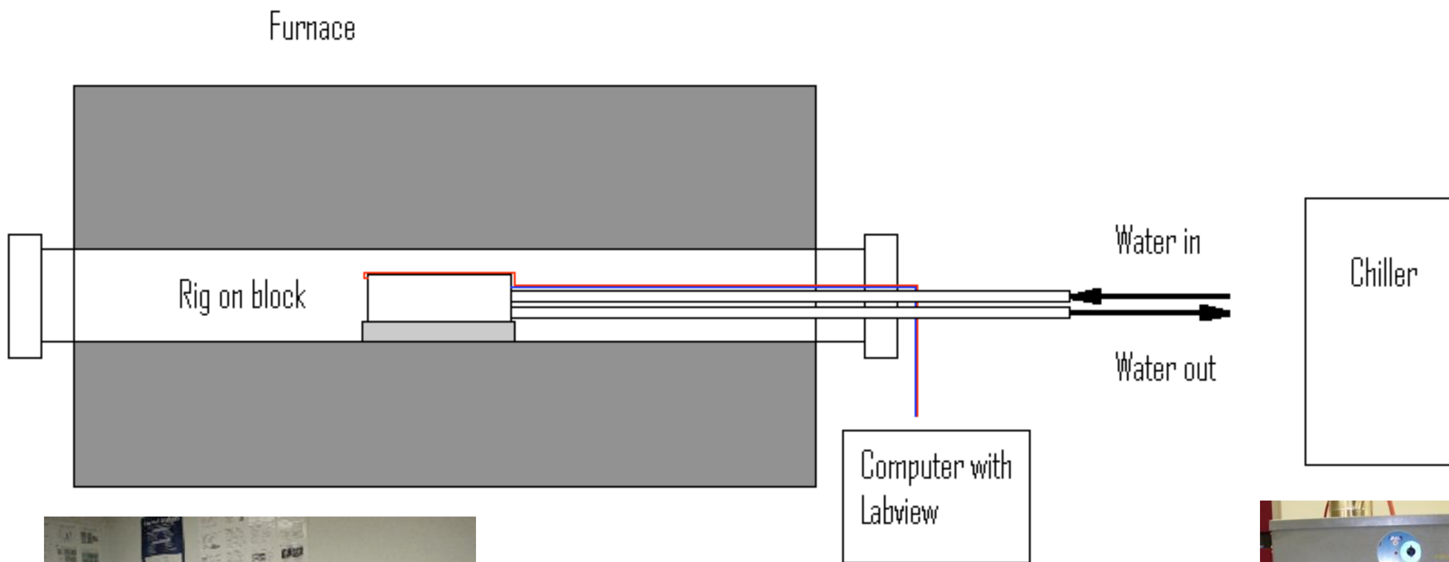
Experimental Apparatus for Thermotransport





Stainless steel inner tube, Copper outer tube joined to a copper plate to draw heat from the sample. Swagelok fittings.

Experimental Thermotransport Work at UCF
Schematic Set-Up (And some pictures)



Temperature (hot- and cold-end) measurements fed into NI controller to regulate water flow that cools the cold-end.

Experimental Diffusion Work at UCF
Parts, Vendors and Status

Part	Status	Vendor
Hot plug (Alumina)	Ordered	Ortech, Inc., Sacramento CA; 916-549-9696; www.ortechceramics.com
Hot-end barrier (Tunsten)	Ordered	Midwest Tungsten Service, Inc. Willowbrook, IL; 630-325-1001; www.tungsten.com
Casing (Alumina)	Ordered	Ortech, Inc., Sacramento CA; 916-549-9696; www.ortechceramics.com
Cold plug (Copper)	New design based on concentric inner/outer tubes with swagelok fitting – no custom production required.	
Measurement/Controller	Received	National Instrument (PXI-8104) 1.86 GHz Celeron M 440 Embedded Controller; http://sine.ni.com/nips/cds/view/p/lang/en/nid/204820
Water chiller	Secured	Used; Haskris R100E Mechanically Refrigerated Closed Circuit Water Chiller
Thermocouple (Type K)	Ordered	Omega Engineering http://www.omega.com/prodinfo/thermocouples.html
Durablanket	Ordered	Thermal Products Company, Clifton Park, NY; http://www.thermalproducts.com
Cu and Stainless steel tubes	Ordered	Onlinemetals Company, Seattle, WA; http://www.onlinemetals.com
Swagelok fittings (Various)	Ordered	Swagelok Company, Solon, OH; http://www.swagelok.com

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Summary

- A diffuse interface model was devised and employed to investigate the effect of thermotransport (a.k.a., thermomigration) process in single-phase and multi-phase alloys of a binary system.
  - ✓ Concentration gradient developed in an initially homogeneous single phase alloy when subjected to temperature gradient.
  - ✓ A and B rich layer formed due to the preferential movement of atoms towards the hot or cold end.

- Experimental work on thermotransport will commence April, 2009. Open to suggestions, collaborations and ideas (e.g., intrinsic frame, alloy systems, assessment/understanding of heat of transport, Q.

Acknowledgements

- Idaho National Laboratory – Drs. Maria Okuniewski, Dennis Keiser, Steven Hayes for Financial Support.
- National Science Foundation Faculty Early Career Development (CAREER) Award (NSF DMR-0238356).
- Dr. Jonathan Guyer and Fipy Development Group, NIST.