NIST Workshop: High Throughput Analysis of Multicomponent Multiphase Diffusion Data April 1-2

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&
Bill Boettinger

Why a Workshop on Diffusion?

• Consensus of NIST Workshop held March 21-22, 2002 *Computational Thermodynamics and Diffusion Modeling*- Promotes continuing interest in thermodynamic databases

• Metallurgy Division participation in DARPA/AIM/GE program on Turbine Disks

• NIST interest in Combinatorial (High Thoughput) Measurement Methods

• Existence of legacy Diffusion in Metals Data base at NIST (J. R. Manning)

Goals

- Improve communication between experts in multicomponent diffusion measurement, analysis and simulation.
- Establish the most efficient method for extracting diffusion data (diffusion coefficients, fluxes, marker location) from multicomponent diffusion couple experiments.
- Provide a forum to solve common diffusion software execution problems.
- Agree on a common diffusion mobility data base assessment procedure.
- Establish a general approach to data handling and diffusion modeling in ordered phases.
- Develop standard problems and web site for inter-laboratory comparison of diffusion simulation methods and data extraction techniques

Agenda

Thursday, April 1, 2004

8:30- 9:00 **Introduction** (Coffee and bagels)

9:00 –10:00 **Review of action items from last workshop**

- Diffusion notation
- Code for $t^{1/2}$ growth for multiple planar layers in binary alloy (Boettinger)
- MultiDiflux (Dayananda)
- Progress on web access to Metallurgy Div. Diffusion Database (Campbell)
- Other

10-10:30 **A new technique to measure ternary interdiffusion coefficients** (Sohn)

10:30-11:00 **Unresolved issues concerning diffusion paths: Horns and the occurrence of type 3 and higher order boundaries** (Morral)

11:00-11:30 **Select ternary multiphase microstructures** (Dayananda)

11:30- 12:00 **Electron probe x-ray microanalysis: doing it right and doing it wrong!** (Newbury)

12:00-1:00 **Lunch**

Agenda

Thursday afternoon

1:00-1:15 **Update on diffusion in unstable phases (bcc Cu)** (Mishin)

1:15-1:45 **Diffusion in ordered phases** (Mishin)

1:45-2:15 **First-principles calculations of the vacancy formation energy** (Wang)

2:15-3:15 **Lateral Deformation of Diffusion Couples (3-D Kirkendall effect)** (Boettinger)

3:15-4:15 Discussion

4:15-4:45 **Implementation of diffusion model for ordered phases** (Campbell)

4:45-5:30 Discussion of diffusion models

6:00 Dinner: Summit Station

Friday April 2

9:00-9:30 Update **Optimization methods** (Campbell and Höglund)
9:30-10:00 Discussion of Standard Problems/ Teaching Methods
10:00-11:00 Open discussion
11:00 –12:00 Action items

Lunch/Adjourn

NIST

Further testing and refinement of database using GE Diffusion Couple Data (FY 2003)

- Binary Couples
  - Single phase couples
    - at 1100 °C for 1000 h : **Ni/Co**
  - Multiphase couples
    - at 1100 °C for 1000 h : Co/Cr, Co/Mo, Co/Nb, Co/W, Cr/Ta, Cr/W, Cr/Mo, Ni/W, Ni/Ta, Ni/Mo, Ni/NiAl(1150 °C)
    - at 850 °C for 4000 h: Ni/W, Co/Fe, Cr/Mo, Cr/Co, Mo/Fe
    - at 700 °C for 4000 h: Fe/Co, Mo/Cr
- Multicomponent Couples
  - Single Phase γ
    - at 1150 °C for 1000 h: René88 /IN718 and Ni/René88
  - γ / γ+γ' or γ+γ' / γ+γ' at 1150 °C for 1000 h

René-95/ René-88	ME3/IN718	IN100/ME3
U720/IN718	IN100/ René-88	René-95/U720
IN718/IN100	U720/ME3	René-95/IN718
ME3/ René-95	ME3/ René-88	IN100/U720

  - γ / B2 or γ+γ' /B2
    - at 1150 °C for 1000 h: NiAl/ René-88, NiAl/Ta
    - at 850 °C for 4000 h: NiAl/ René-88, NiAl/Ta
  - TCP Couples: (Rene88-X)
    - at 1150 °C for 1000 h: X= Ta, W
    - at 850 °C for 4000 h: X=Ta, W, Co, Cr, Fe, Mo, Ni, Ti
    - at 700 °C for 4000 h: X=Co, Cr, Fe, Mo

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NIST participation in GE-AIM (DARPA) Program
γ' Precipitation Model

Experimental diffusion data
(Binary data)
J.C. Zhao, GE-CRD
NIST Diffusion Data Center

Multicomponent diffusion data
B. Mueller, Howmet
J-C. Zhao, GE-CRD

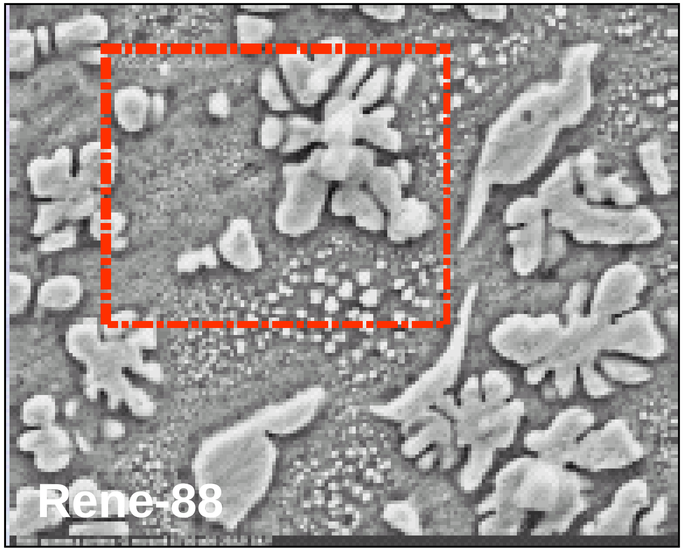
Diffusion Mobility Database
NIST

Thermodynamic Databases
Thermotech, UW, NIST

Lattice parameters
Questek

γ' precipitation
Questek, GE

Models for mechanical properties
GE



Linear cooling rate 0.2 °C/s

NIST also providing guidance on γ' precipitation modeling, simple coding for thermodynamic calculations, modeling interfacial energies.

Multicomponent Mobility Database for FCC phase of Superalloys

Campbell, Boettinger & Kattner, Acta Mat.50 (2002) 775-792.

Ni = solvent

Reduced (n-1)Diffusion Matrix at 1293 °C

René-N4 (x10^{-14} m^2/s)

	Al	*Co*	*Cr*	*Mo*	*Nb*	*Ta*	*Ti*	*W*
Al	+119.5	+13.93	+34.83	+34.34	+42.43	+51.50	+49.51	+53.22
Co	−11.37	+17.00	−8.25	−5.67	−5.55	−1.83	−7.10	−9.69
Cr	−4.26	−5.37	+13.67	−3.21	+8.93	+9.91	+8.25	+2.49
Mo	−8.33	−0.280	−0.426	+7.57	−0.55	−0.36	−0.17	−0.45
Nb	+0.31	+0.25	+0.66	+0.27	+24.05	+0.74	+0.85	+0.31
Ta	−0.68	+0.33	+0.53	+0.24	+0.26	+0.76	+0.50	+0.23
Ti	+1.63	+1.35	+4.94	+4.94	+6.25	+6.57	+23.62	+5.41
W	−1.81	−0.62	−0.55	−0.60	−1.22	−0.83	−0.70	+3.40

René-N5 (x10^{-14} m^2/s)

	Al	*Co*	*Cr*	*Hf*	*Mo*	*Re*	*Ta*	*W*
Al	+93.16	+13.92	+33.46	−6.51	+33.42	25.44	+48.63	+50.87
Co	−6.51	+27.22	−8.56	−27.64	−4.95	−5.11	+3.87	−9.21
Cr	+4.15	−4.23	+21.02	−6.25	−0.22	−0.78	+13.81	+6.89
Hf	0.86	+0.07	+1.70	+262.1	+1.52	0.87	+2.37	+1.84
Mo	−0.35	−0.30	−0.30	−1.905	+7.71	−0.25	−0.13	−0.19
Re	−0.75	−0.32	−0.36	−2.59	−0.25	+0.08	−0.51	−0.32
Ta	−0.03	+0.33	+0.98	−4.17	+0.64	+0.86	+7.75	+0.87
W	−1.18	−0.57	−0.54	−4.51	−0.39	−0.11	−0.76	+0.59

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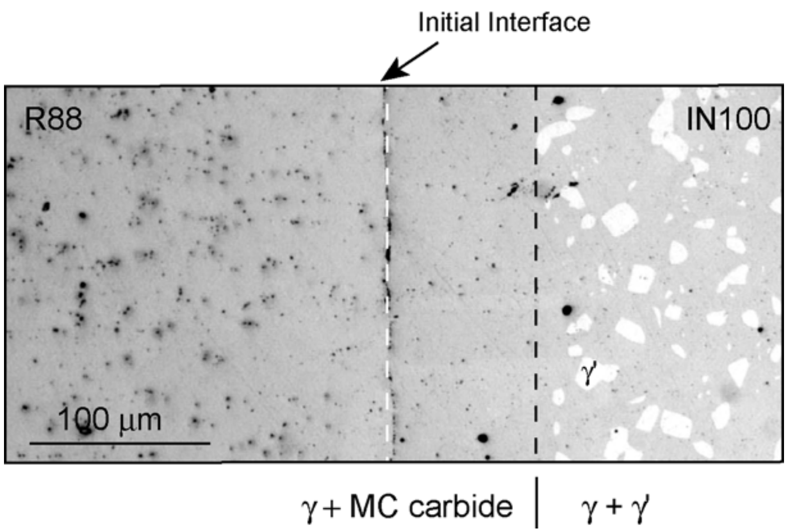
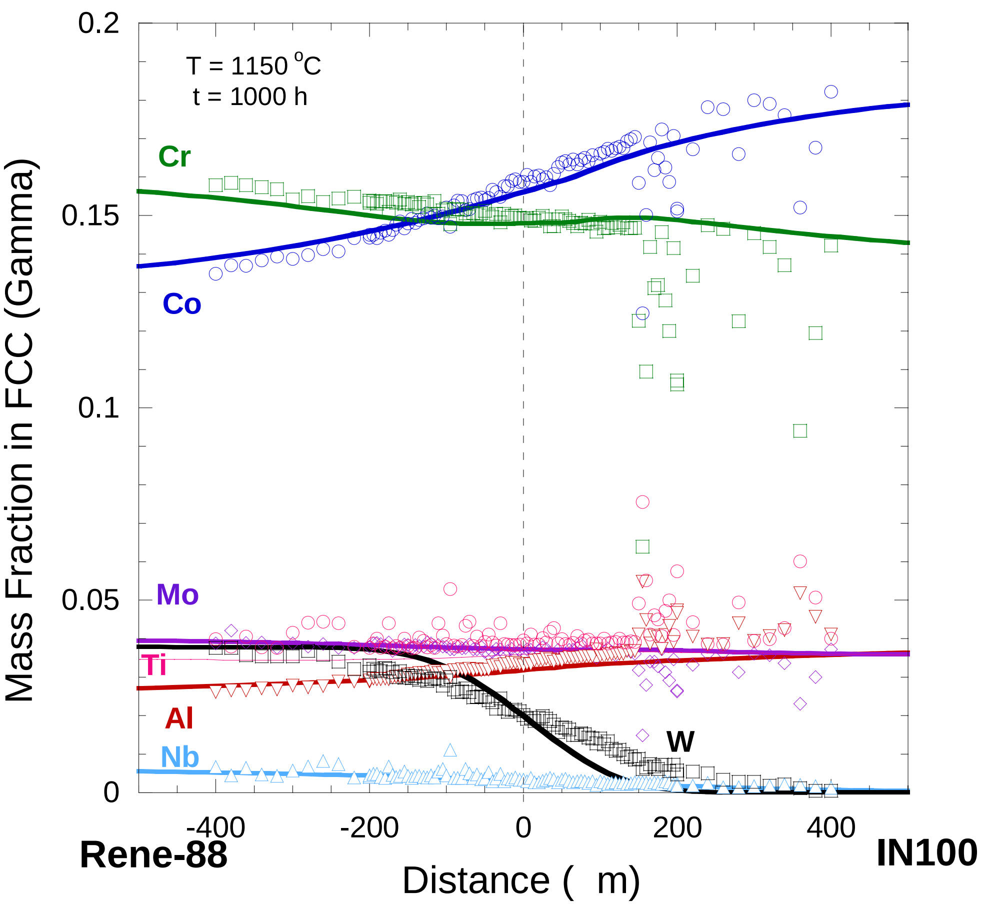
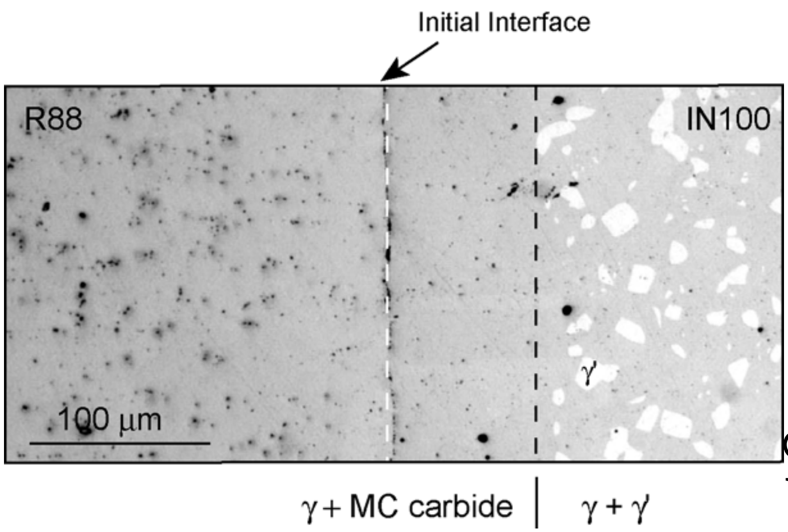
René-88/IN-100; 1000 h at 1150 °C

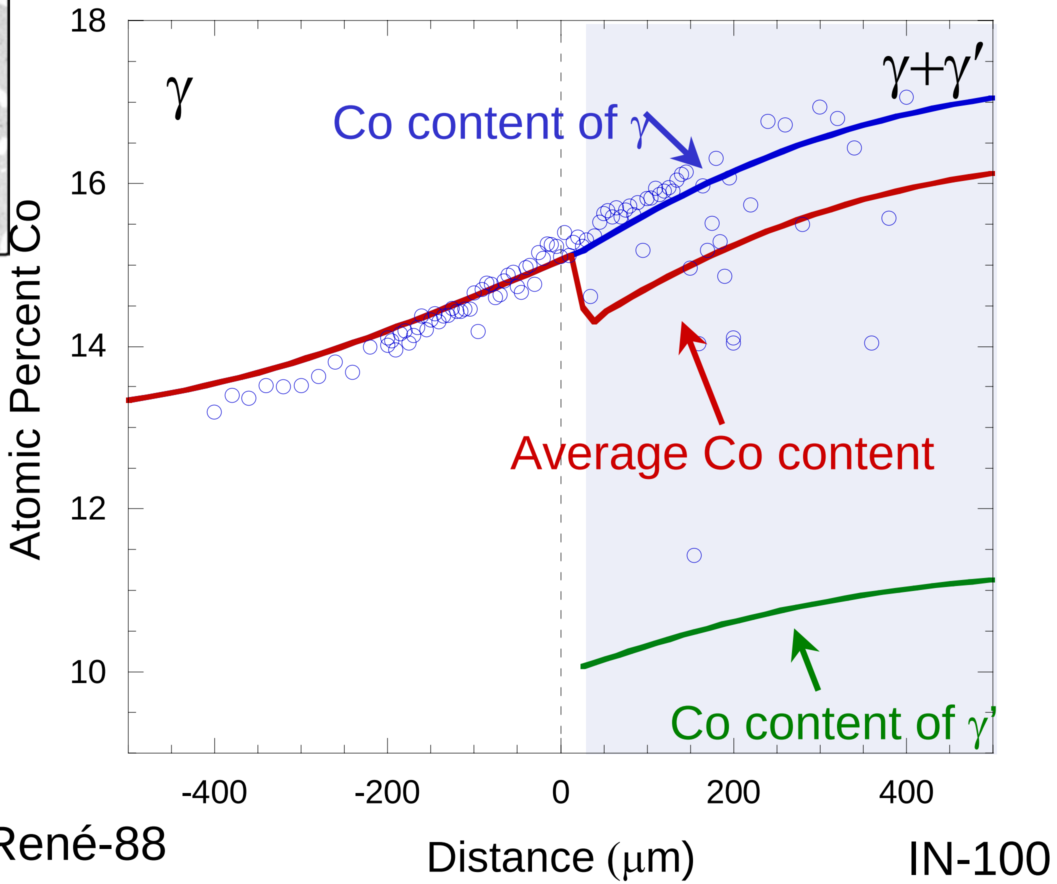




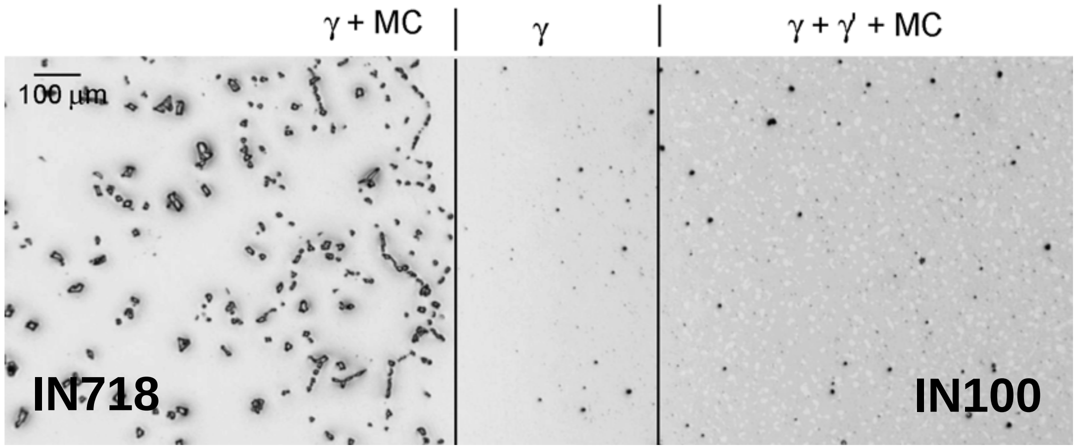
Experimental data from J-C. Zhao, GE Global Research

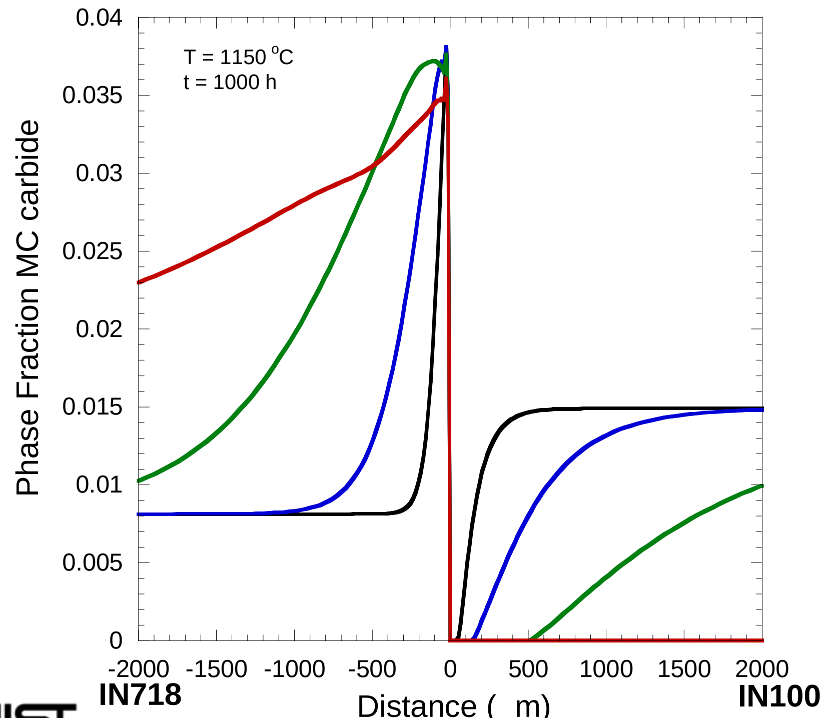
René-88/IN-100; 1000 h at 1150 °C

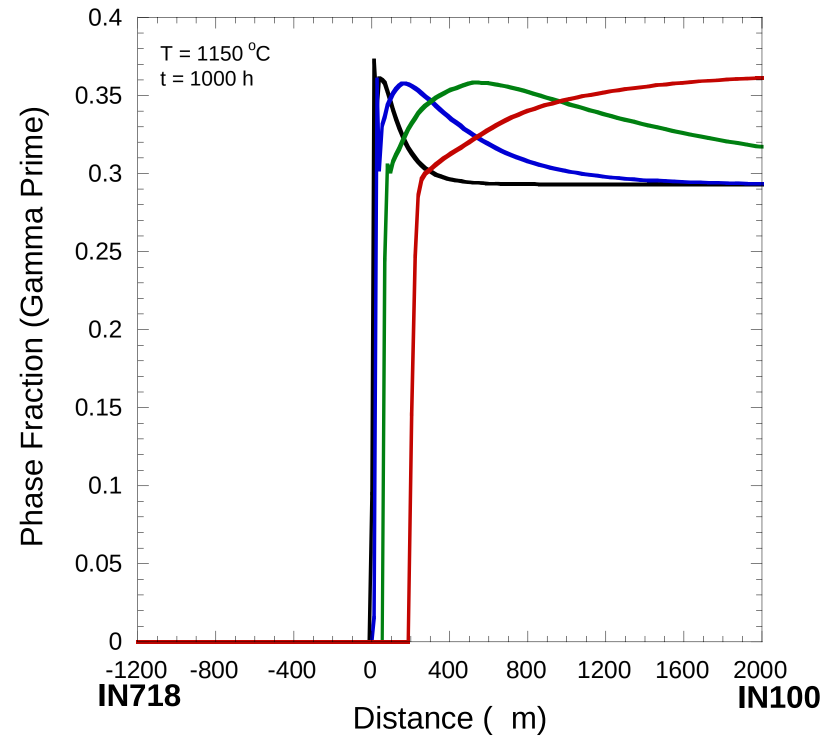




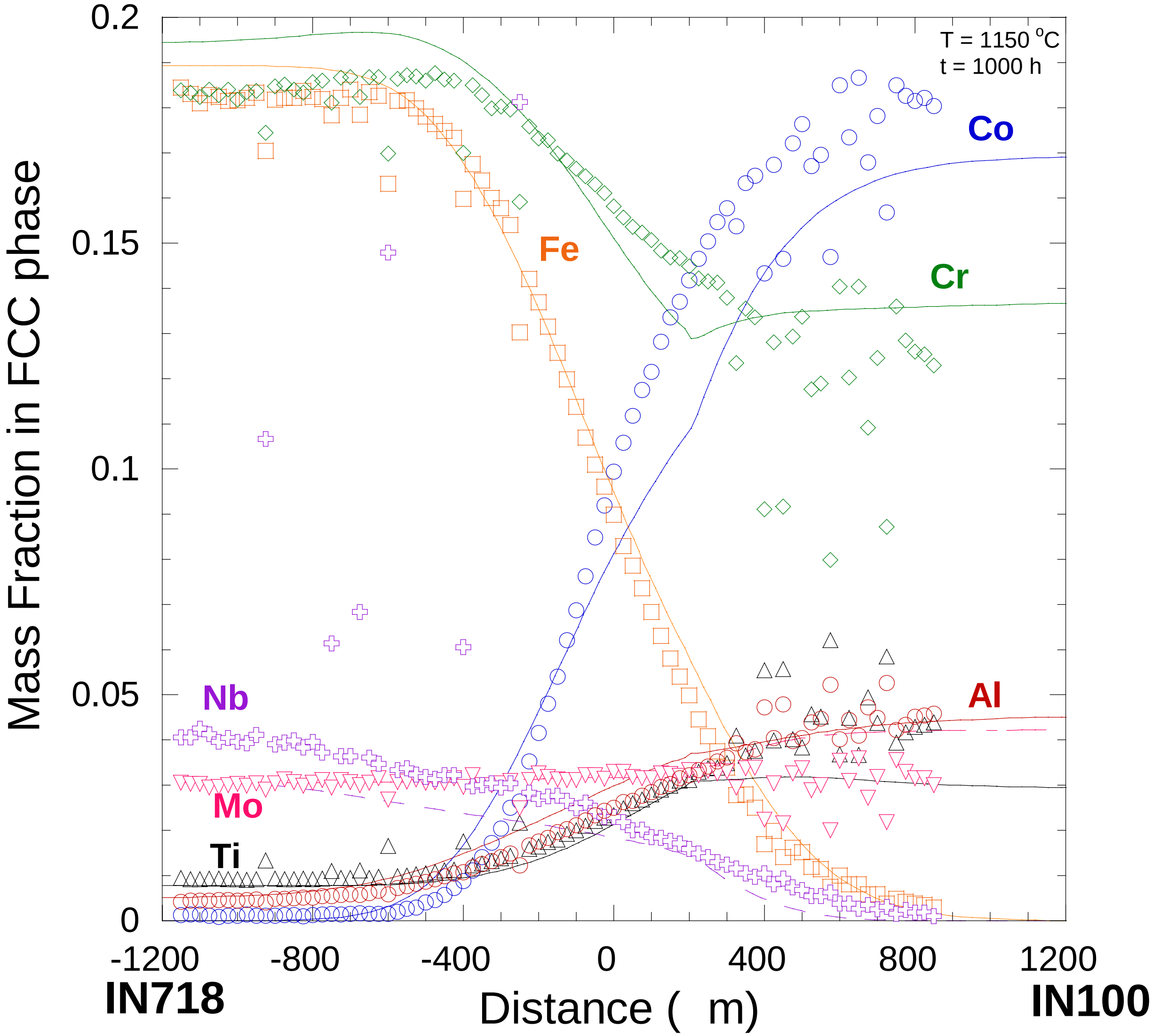
IN-718/IN-100; 1000 h at 1150 °C







IN-718/IN-100; 1000 h at 1150 °C



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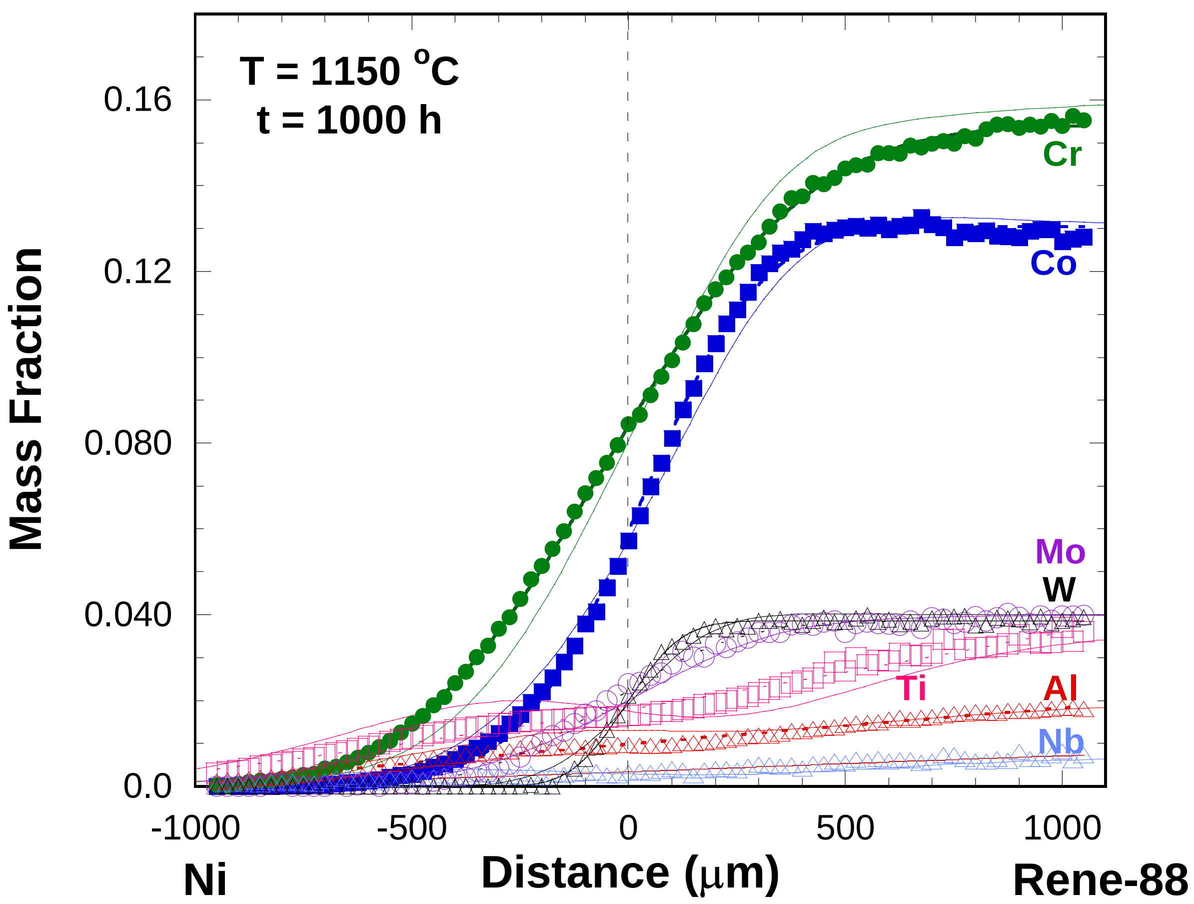
Ni-René88



Symbols = GE experimental data
Solid lines = NIST Database/DICTRA prediction
Dashed lines = Error function fit

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Definitions

Coefficient	General Notation	DICTRA notation
Tracer Diffusivity	$D_i^* = \tilde{\nu}\beta a^2 f \exp\left(\frac{\Delta S_{Va}^f + \Delta S_{Va}^m}{k}\right)\exp\left(-\frac{\Delta H_{Va}^f + \Delta H_{Va}^m}{kT}\right)$ $\tilde{\upsilon}$ = vibration frequency a = lattice parameter $\beta = 1$ for FCC and BCC and 1/8 for diamond cubic f = correlation factor $D = D_0 \exp\left(\frac{-Q}{RT}\right)$	$\boxed{D_k^* = RTM_k}$ $M_k = \delta^2 \nu \exp\left(-\frac{\Delta G_{kVa}^*}{RT}\right)\frac{1}{RT}$
Intrinsic Diffusivity (partial chemical)	$D_i = D_i^*\left[1 + \frac{\partial \log \gamma}{\partial \log N}\right]$	$\boxed{{}^{i}D_{kj} = c_k M_{kVa} \frac{\partial \mu_k}{\partial c_j}}$
Chemical Diffusivity (Interdiffusion)	$\tilde{D} = x_A D_B + x_B D_A$ (binary) D_i and $\tilde{D}$ are related by the velocity of Kirkendall frame, $v = -J_{Va}V_M$	$\boxed{D_{kj} = \sum_{i=1}^{n}(\delta_{ik} - x_k)x_i M_i \frac{\partial \mu_i}{\partial x_j} V_m}$

Ternary A-B-C System

In the lattice fixed frame of reference, assuming a substitutional solid solution for a given phase:

$$M_{kVa} = M_k^0 \exp\left(\frac{-\Delta G_{kVa}}{RT}\right)\frac{1}{RT}$$

Assume: $M_k^0 = 1$

Then the activation energy, Q_k:

$$\Delta G^*_{AVa} = x_A y_{Va} \Delta G_A^{A:Va} + x_B y_{Va} \Delta G_A^{B:Va} + x_C y_{Va} \Delta G_A^{C:Va} + \Delta G^{excess}$$
$$\Delta G^*_{BVa} = x_A y_{Va} \Delta G_B^{A:Va} + x_B y_{Va} \Delta G_B^{B:Va} + x_C y_{Va} \Delta G_B^{C:Va} + \Delta G^{excess}$$
$$\Delta G^*_{CVa} = x_A y_{Va} \Delta G_C^{A:Va} + x_B y_{Va} \Delta G_C^{B:Va} + x_C y_{Va} \Delta G_C^{C:Va} + \Delta G^{excess}$$

Note x_i = mole fraction of component i
y_i = site fraction of component i
on a given sublattice

Since this is a substitutional solid solution with no interstitials, $y_{Va} = 1$:

Concentration variables: $c_k = \frac{x_k}{V_m} = \frac{x_k}{\sum_{j=1}^{n} x_j V_j}$ *and* $V_j = \left(\frac{\partial V}{\partial N_j}\right)_{P,T,N_k}$

Where x_k is the mole fraction of component k, V_j is the partial molar volume and N_j is the number of moles of component k.

Assume all the substitutional components have the same partial molar volume: $V_j = V_s$ then for the A-B-C system: $V_m = x_A V_s + x_B V_s + x_C V_s$

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Ternary A-B-C System

In the lattice fixed frame of reference: $\sum_{k=1}^{n} J_k = -J_{Va} = J_A + J_B + J_C$

$$J_k = -\sum_{j=1}^{n} {}^iD_{kj} \frac{\partial c_j}{\partial z} = -\sum_{j=1}^{n-1} L_{kk} \frac{\partial \mu_k}{\partial c_j} \frac{\partial c_j}{\partial z}$$

$${}^iD_{kj} = L_{kk} \frac{\partial \mu_k}{\partial c_j} = c_k y_{Va} M_{kVa} \frac{\partial \mu_k}{\partial c_j}$$

Note y_{Va} =1 and for simplicity drop the *Va* from $M_{kVa} = M_k$

$$L_{AA} = x_A M_A \qquad c_i = \frac{x_i}{V_m}$$

$$L_{BB} = x_B M_B$$

$$L_{CC} = x_C M_C$$

$${}^iD_{AA} = L_{AA} \frac{\partial \mu_A}{\partial c_A} \qquad {}^iD_{AB} = L_{AA} \frac{\partial \mu_A}{\partial c_B} \qquad {}^iD_{AC} = L_{AA} \frac{\partial \mu_A}{\partial c_C}$$

$${}^iD_{BA} = L_{BB} \frac{\partial \mu_B}{\partial c_A} \qquad {}^iD_{BB} = L_{BB} \frac{\partial \mu_B}{\partial c_B} \qquad {}^iD_{BC} = L_{BB} \frac{\partial \mu_B}{\partial c_C}$$

$${}^iD_{CA} = L_{CC} \frac{\partial \mu_C}{\partial c_A} \qquad {}^iD_{CB} = L_{CC} \frac{\partial \mu_C}{\partial c_B} \qquad {}^iD_{CC} = L_{CC} \frac{\partial \mu_C}{\partial c_C}$$

Ternary A-B-C System

In the volume-fixed frame of reference: $\sum_{k=1}^{n} J_k V_k = 0 = (J_A + J_B + J_C)\cdot V_s$

$$J_k = -\sum_{i=1}^{n} L'_{ki}\frac{\partial \mu_i}{\partial z} = -\sum_{i=1}^{n} L'_{ki}\sum_{j=1}^{n}\frac{\partial \mu_i}{\partial c_j}\frac{\partial c_j}{\partial z} = -\sum_{j=1}^{n} D_{kj}\frac{\partial c_j}{\partial z}$$

$$\boxed{D_{kj} = -\sum_{i=1}^{n} L'_{ki}\frac{\partial \mu_i}{\partial c_j} = \sum_{i=1}^{n}(\delta_{ik} - x_k)x_i M_i \frac{\partial \mu_i}{\partial x_j}V_m}$$

See next page for expansion

$$L'_{kj} = \sum_{i=1}^{n}\left[\delta_{ik} - x_k\left(\frac{V_i}{V_m}\right)\right]L_{ij} \qquad L_{kk} = c_k y_{Va} M_{kVa}$$

Equivalent forms

Or $J_k = -\sum_{i=1}^{n} L''_{ki}\left[\nabla\mu_i - \left(\frac{V_i}{V_m}\right)\nabla\mu_n\right]$

$$L''_{ki} = \sum_{j=1}^{n}\left[\delta_{ij} - x_i\left(\frac{V_i}{V_m}\right)\right]L'_{kj} = \sum_{j=1}^{n}\sum_{r=1}^{n}\left[\delta_{ir} - x_i\left(\frac{V_r}{V_m}\right)\right]\left[\delta_{jk} - x_k\left(\frac{V_j}{V_m}\right)\right]L_{jr}$$

$$\boxed{D_{kj} = \sum_{i=1}^{n} L''_{ki}\frac{\partial[\mu_i - (V_i/V_m)\mu_n]}{\partial c_j}} = \sum_{i=1}^{n} L'_{ki}\frac{\partial \mu_i}{\partial c_j} - \sum_{i=1}^{n} x_i\frac{\partial \mu_i}{\partial c_j}\sum_{r=1}^{n}\left(\frac{V_r}{V_m}\right)L'_{kr}$$

Recall that the Gibbs–Duhem equations provides that: $\sum_{i=1}^{n} x_i\frac{\partial \mu_i}{\partial c_j} = 0$ thus, $D_{kj} = \sum_{i=1}^{n} L'_{ki}\frac{\partial \mu_i}{\partial c_j}$

Reduce diffusivities when $V_k = V_s$: $D^n_{kj} = D_{kj} - D_{kn} \qquad J_k = -\sum_{j=1}^{n-1} D^n_{kj}\frac{\partial c_j}{\partial z}$

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Ternary A-B-C

$$D_{kj} = -\sum_{i=1}^{n} L'_{ki} \frac{\partial \mu_i}{\partial c_j} = \sum_{i=1}^{n} \left(\delta_{ik} - x_k\right) x_i M_i \frac{\partial \mu_i}{\partial x_j} V_m$$

$$D_{AA} = (1 - x_A) x_A M_A \frac{\partial \mu_A}{\partial x_A} V_m + (0 - x_A) x_B M_B \frac{\partial \mu_B}{\partial x_A} V_m + (0 - x_A) x_C M_C \frac{\partial \mu_C}{\partial x_A} V_m$$

$$D_{AB} = (1 - x_A) x_A M_A \frac{\partial \mu_A}{\partial x_B} V_m + (0 - x_A) x_B M_B \frac{\partial \mu_B}{\partial x_B} V_m + (0 - x_A) x_C M_C \frac{\partial \mu_C}{\partial x_B} V_m$$

$$D_{AC} = (1 - x_A) x_A M_A \frac{\partial \mu_A}{\partial x_C} V_m + (0 - x_A) x_B M_B \frac{\partial \mu_B}{\partial x_C} V_m + (0 - x_A) x_C M_C \frac{\partial \mu_C}{\partial x_C} V_m$$

$$D_{BA} = (0 - x_B) x_A M_A \frac{\partial \mu_A}{\partial x_A} V_m + (1 - x_B) x_B M_B \frac{\partial \mu_B}{\partial x_A} V_m + (0 - x_B) x_C M_C \frac{\partial \mu_C}{\partial x_A} V_m$$

$$D_{BB} = (0 - x_B) x_A M_A \frac{\partial \mu_A}{\partial x_B} V_m + (1 - x_B) x_B M_B \frac{\partial \mu_B}{\partial x_B} V_m + (0 - x_B) x_C M_C \frac{\partial \mu_C}{\partial x_B} V_m$$

$$D_{BC} = (0 - x_B) x_A M_A \frac{\partial \mu_A}{\partial x_C} V_m + (1 - x_B) x_B M_B \frac{\partial \mu_B}{\partial x_C} V_m + (0 - x_B) x_C M_C \frac{\partial \mu_C}{\partial x_C} V_m$$

$$D_{CA} = (0 - x_C) x_A M_A \frac{\partial \mu_A}{\partial x_A} V_m + (0 - x_C) x_B M_B \frac{\partial \mu_B}{\partial x_A} V_m + (1 - x_C) x_C M_C \frac{\partial \mu_C}{\partial x_A} V_m$$

$$D_{CB} = (1 - x_C) x_A M_A \frac{\partial \mu_A}{\partial x_B} V_m + (0 - x_C) x_B M_B \frac{\partial \mu_B}{\partial x_B} V_m + (1 - x_C) x_C M_C \frac{\partial \mu_C}{\partial x_B} V_m$$

$$D_{CC} = (1 - x_C) x_A M_A \frac{\partial \mu_A}{\partial x_C} V_m + (0 - x_C) x_B M_B \frac{\partial \mu_B}{\partial x_C} V_m + (1 - x_C) x_C M_C \frac{\partial \mu_C}{\partial x_C} V_m$$

Relation between Intrinsic and Interdiffusion Coefficients

From Sohn and Dayananda (*Met. Mat. Trans* **33A** (2002) 3375)

Published notation:

$$\tilde{D}_{ij}^{n} = D_{ij}^{n} - N_i \sum_{k=1}^{n} D_{kj}^{n}$$

$\tilde{D}_{ij}^{n} =$ interdiffusion coefficient

$D_{ij}^{n} =$ intrinsic diffusion coefficient (reduced)

$N_i =$ atom fraction

DICTRA notation:

$$\tilde{D}_{ij}^{n} = {}^{i}D_{ij}^{n} - x_i \sum_{k=1}^{n} {}^{i}D_{kj}^{n}$$

Example: $\tilde{D}_{AA}^{C} = {}^{i}D_{AA}^{C} - x_A\left({}^{i}D_{AA}^{C} + {}^{i}D_{BA}^{C} + {}^{i}D_{CA}^{C}\right)$

$\tilde{D}_{AA}^{C} = D_{AA} - D_{AC}$ (see previous slide for expansion of D_{AA} and D_{AC})

$$\tilde{D}_{AA}^{C} = x_A V_m\left[(1-x_A)M_A\frac{\partial\mu_A}{\partial x_A} - x_B M_B\frac{\partial\mu_B}{\partial x_A} - x_C M_C\frac{\partial\mu_C}{\partial x_A}\right] - x_A V_m\left[(1-x_A)M_A\frac{\partial\mu_A}{\partial x_C} - x_B M_B\frac{\partial\mu_B}{\partial x_C} - x_C M_C\frac{\partial\mu_C}{\partial x_C}\right]$$

Note $\quad {}^{i}D_{kj} = x_k M_k \dfrac{\partial\mu_k}{\partial x_j}$

$$\tilde{D}_{AA}^{C} = x_A V_m\left[M_A\frac{\partial\mu_A}{\partial x_A} - {}^{i}D_{AA} - {}^{i}D_{BA} - {}^{i}D_{CA}\right] - x_A V_m\left[M_A\frac{\partial\mu_A}{\partial x_C} - {}^{i}D_{AC} - {}^{i}D_{BC} - {}^{i}D_{CC}\right]$$

$$\tilde{D}_{AA}^{C} = x_A V_m M_A\frac{\partial\mu_A}{\partial x_A} - x_A V_m M_A\frac{\partial\mu_A}{\partial x_C} - x_A V_m\left[\left({}^{i}D_{AA} - {}^{i}D_{AC}\right) + \left({}^{i}D_{BA} - {}^{i}D_{BC}\right) + \left({}^{i}D_{CA} - {}^{i}D_{CC}\right)\right]$$

$$\tilde{D}_{AA}^{C} = \left({}^{i}D_{AA} - {}^{i}D_{AC}\right) - x_A V_m\left[\left({}^{i}D_{AA} - {}^{i}D_{AC}\right) + \left({}^{i}D_{BA} - {}^{i}D_{BC}\right) + \left({}^{i}D_{CA} - {}^{i}D_{CC}\right)\right]$$

Rewrite in terms of reduce diffusivities

$$\tilde{D}_{AA}^{C} = {}^{i}D_{AA}^{C} - x_A V_m\left[{}^{i}D_{AA}^{C} + {}^{i}D_{BA}^{C} + {}^{i}D_{CA}^{C}\right]$$

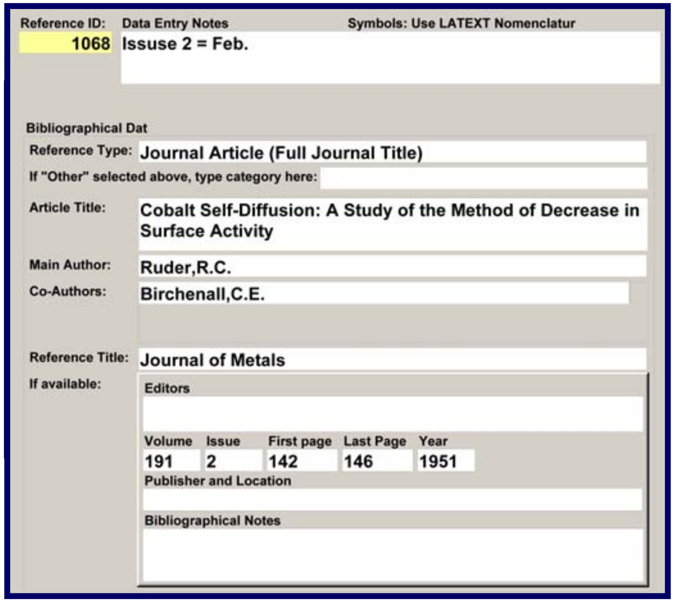
Note:

$${}^{i}D_{kj}^{n} = {}^{i}D_{kj} - {}^{i}D_{kn}$$
$${}^{i}D_{AA}^{C} = {}^{i}D_{AA} - {}^{i}D_{AC}$$
$${}^{i}D_{BA}^{C} = {}^{i}D_{BA} - {}^{i}D_{BC}$$
$${}^{i}D_{CA}^{C} = {}^{i}D_{CA} - {}^{i}D_{CC}$$

Diffusion Database Center

C. E. Campbell, U.R. Kattner, C. Beauchamp, K. Dotterer, H. Gates, S. Tobery

- **Goal:** To make the NIST paper-based diffusion database center publicly available.
  - Convert to a searchable electronic form to be access over the internet
- **Motivation**
  - Industrial and academic support: GE $5K initiation
  - Center represents an unique collection summarizing the diffusion work between 1965-1980
- **Task:**
  - Need to enter 25000 bibliographic and diffusion system cards
  - Convert paper documents to electronic documents
  - Develop searchable database
- **Accomplishments (2003)**
  - Developed database entry strategy
  - Entered 6000 bibliographic cards
  - Purchase high speed scanner and software


Reference ID: 1068
Data Entry Notes: Issuse 2 = Feb.
Symbols: Use LATEXT Nomenclatur
Bibliographical Dat
Reference Type: Journal Article (Full Journal Title)
If "Other" selected above, type category here:
Article Title: Cobalt Self-Diffusion: A Study of the Method of Decrease in Surface Activity
Main Author: Ruder,R.C.
Co-Authors: Birchenall,C.E.
Reference Title: Journal of Metals
If available:
Editors
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NIST

Upcoming Events

- DIMAT 2004 (July 18-24, 2004, Poland)
  - http://www.dimat2004.agh.edu.pl

- First International Conference on Diffusion in Solids and Liquids
  - July 6-8,2005, Aveiro, Portugal
  - http://event.ua.pt/dsl2005/index.html
  - Abstracts due Dec. 31, 2004

TMS Annual Meeting Feb., 2005

Multicomponent Multiphase Diffusion Symposium in Honor of John Morral

Symposium abstract: Throughout his career, John Morral has dedicated his work to the understanding and application of multicomponent diffusion. This symposium, in honor of John Morral's 65th birthday, will highlight both experimental and theoretical work in a variety of multicomponent multiphase diffusion problems. This work is increasingly important in improving industrial materials processing and development. In addition to invited speakers, contributed papers on mutlicomponent diffusion and microstructure evolution are solicited. Highlighted topics are to include diffusion kinetics of high temperature coatings and processing, 'zig-zag' diffusion paths, internal oxidation, carburizing, nitriding, and alloy heat treatment.

Abstract deadline: August 1, 2004

Organizers:

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