УДК 539.219.3 PACS number 66.30.-h, 02.30.Jr

Bartłomiej Wierzba and Marek Danielewski

Interdisciplinary Centre for Materials Modelling, FMS&C, AGH University of Science and Technology,
Al. Mickiewicza 30, 30-059 Cracow, Poland
bwierzba@agh.edu.pl

STOCHASTIC SEARCHING OF DIFFUSIVITY IN MULTICOMPONENT ALLOYS BY HIERARCHICAL GENETIC STRATEGY

Diffusivities in the multicomponent solutions are determined using the inverse diffusion problem. Such solutions are non-unique and computed diffusivities can not be treated as genuine material constants. In this work the Hierarchical Genetic Strategy with real number encoding (HGS-FP) is combined with the Darken method. The HGS-FP method is used to approximate the variable (effective) intrinsic diffusivities in alloys at 1273 K. It will be shown that the Redlich-Kister approximation of the composition dependent diffusivities gives satisfactory agreement with experimental data. The presented calculations show a great future potential of HGS-FP strategy. The parallel version of HGS allows decreasing the computation time.

Коефіцієнти дифузії в багатокомпонентній системі визначаються в рамках обереної задачі дифузії. Такі розв'язки не є єдиними і тому не можуть трактуватися як матеріальні константи. В даній роботі генетичий алгоритм поєднаний із методом Даркена і застосовується для визначення парціальних коефіцієнтів дифузії в сплавах Fe-Ni-Cu при 1273 K. Показано, що наближення Редліха-Кістера для концентраційної залежності коефіцієнтів дифузії задовільно узгоджується з експериментальними даними. Представлені розрахунки демонструють великий потенціал генетичних алгоритмів. Паралельна версія алгоритму дозволяє зменшити час розрахунків.

Коэффициенты диффузии в многокомпонентной системе определяются в пределах обратной задачи диффузии. Такие решения не единственны и поэтому не могут интерпретироваться как материальные константы. В этой работе генетический алгоритм сочетается с методом Даркена и используется для определения парциальных коэффициентов диффузии в сплавах Fe-Ni-Cu при 1273 K. Показано, что приближение Редлиха-Кистера для концентрационной зависимости коэффициентов диффузии удовлетворительно согласуется с экспериментальными данными. Представленые расчёты демонстрируют большой потенциал генетических алгоритмов. Паралельная версия алгоритма разрешает уменшить время расчётов.

Keywords: *optimization, genetic algorithm, hierarchical genetic strategy with real number encoding, interdiffusion, simulation.*

Introduction

The applicability of the optimization techniques are of the interest in many researches. The genetic algorithms are of the highest efficiency in the solving problems with many optima [1,2]. The Hierarchical Genetic Strategy (HGS) introduced in 2000 by Kołodziej and Schaefer [3] and further generalized by Wierzba et al. [4] by introducing the floating point encoding is one of such efficient algorithms. Its accuracy depends on the method of defined genetic spaces, where at the lowest ones the searching of the extreme is more chaotic than on the higher ones. The

introduction of all the genetic operators in floating point representation like mutation and crossover improves the efficiency of the optimization strategy. The HGS accuracy and low computational cost for multimodal benchmarks was shown in [3].

The Boltzmann-Matano analysis is widely used to calculate the diffusion coefficients in binary diffusion couples. Some modifications of the method to calculate the diffusion coefficients in more complex systems are also known [5].

In this paper the Hierarchical Genetic Strategy with real number encoding (HGS-FP) with the Darken method is combined to calculate the intrinsic diffusion coefficients in Cu-Fe-Ni alloy at 1273 K. It will be shown that the Redlich-Kister approximation of the composition dependent diffusivities gives satisfactory agreement with experimental data. Moreover, the HGS strategy is an efficient tool for finding the physico-chemical parameters.

1. Darken's method.

In this paper the inverse method to obtain the diffusion parameters which allow simulating the interdiffusion processes in multicomponent mixtures is shown. This method is based on Darken's [6] concept of interdiffusion and its generalization [7]. The main equation that describes Darken's concept is mass conservation law:

$$\frac{\partial c_i}{\partial t} = -\frac{\partial}{\partial x}\left(c_i \upsilon_i^d + c_i \upsilon^D\right), \qquad i = 1,...,r \tag{1}$$

where: c_i denotes the molar concentration, and υ_i^d, υ^D are the diffusion velocity of the
i-th component and Darken velocity (drift), respectively. The diffusion velocity of *i*-th component can be introduced by Nernst-Planck [8, 9] equation:

$$\upsilon_i^d = B_i F_i = -B_i \operatorname{grad} \mu_i^{ch}, \qquad i = 1,...,r \tag{2}$$

where: B_i and F_i denote the mobility, $B_i = D_i^* / RT$, and force acting on the *i*-th component. The forces are equal to the chemical potential gradient, μ_i^{ch}.

Darken velocity is a common velocity in the alloy (common for every component) due to the unbalanced diffusion fluxes [10]:

$$\upsilon^D = -\sum_{i=1}^{r} \Omega_i c_i \upsilon_i^d \tag{3}$$

The boundary conditions for a closed system (not exchanging mass with surrounding) are given by:

$$J_i^d\left(\pm\Lambda, t\right) = 0, \qquad i = 1,...,r \tag{4}$$

where J_i^d denote the diffusion flux, and equal $J_i^d = c_i \upsilon_i^d$.

The initial conditions can be introduced by any set of points (e.g. the hevyside function)

$$c_i\left(x, 0\right) = c_i^0, \qquad i = 1,...,r \tag{5}$$

2. Diffusivities.

In this work the Redlich-Kister approximation of interdiffusion coefficients [11] was applied. The approximation is based on the CALPHAD approach, which allows us to calculate the composition-dependent intrinsic diffusion coefficients from the known self-diffusion coefficients. The self diffusion coefficients can be expressed by:

$$D_i^* = D_i^0 \exp\left(-\frac{Q_i}{RT}\right), \qquad i = 1,2,...,r \tag{6}$$

where: D_i^0 and Q_i denote the pre-exponential constant and activation energy, respectively.

Following DICTRA [12], in the lines of the Calphad approach, the composition dependence of these two factors in alloys (composition dependence of the intrinsic diffusivities), is represented with a linear combination of the values at each endpoint of the composition space. Using the Redlich-Kister expansion and Eq. (6) one gets:

$$\Phi^i = RT \ln D_i^* = RT \ln D_i^0 - Q_i, \qquad i = 1,2,...,r \tag{7}$$

where: Φ^i can be interpreted as free energy of self-diffusion of the *i*-th component.

The free energy of intrinsic diffusion coefficient in *2*-component mixture (solid solution) is computed using the following relation [12]:

$$\Phi_l = \sum_{j=1}^{r} N_j \Phi_l^j + \sum_{j=1}^{r} \sum_{k>j}^{r} N_j N_k \left[\sum_{p=0}^{m} {}^p\Phi_l^{j,k} \left(N_j - N_k\right)^p \right], \qquad l = 1,2,...,r \tag{8}$$

where: Φ_l can be interpreted as free energy of intrinsic diffusion in the multicomponent alloy and ${}^p\Phi_l^{j,k}$ are binary interaction parameters.

Thus, from the Eq. (8), the intrinsic diffusion coefficient in the alloy is given by:

$$D_l = \exp\left(\frac{\Phi_l}{RT}\right), \qquad l = 1,2,...,r\,. \tag{9}$$

3. Hierarchical Genetic Strategy with real number encoding (HGS-FP) [13].

The efficiency, i.e. the low computational cost and effectiveness in finding global optima (maxima or minima) of HGS-FP, comes from concurrent search in the optimization space by many small populations of individuals (solutions). The creation of these populations is governed by genetic processes with low complexity. The main engine of the HGS-FP, in distinction to majority of genetic algorithms, is the mechanism that is based on the real-number encoding. The genetic operators are: 1) the mutation (i.e. the genotype perturbation of the “parent” individual) which allows random relocation of the individuals in the environment (optimization space) and 2) the arithmetic crossover, which is quasi-deterministic combination of the “parents” genotypes. The real number encoding used in HGS-FP is much more efficient than usually used binary encodings (0,1 codes). Its efficiency comes from the conservation of the natural (topological) space where all variables are real numbers.

3.1. Structure of HGS-FP

The main engine of the Hierarchical Genetic Strategy with real number encoding is running a set of evolutionary processes [14,15]. The algorithm in each evolving step creates new population which is searched for optima with higher precision expressed by real numbers.

Let $\Im = [a,b]^N \subset \mathbf{R}^N$ denote the fitness function domain (goal function domain). *N* is the dimension of the problem (number of variables) and *a, b* are the left and the right boundary of the searching space (goal function domain or optimization space), respectively. The two genetic operators for floating point representation are given by:

1) crossover (generation of the new individual between two already existing ones):

$$Y_i = X_i^1 + \mathcal{N}(mean,\sigma)(X_i^2 - X_i^1), \quad i = 1,...,N \tag{10}$$

2) mutation (generation of the new individual basing on the already existing one):

$$Y_i = X_i^1 + (\mathcal{N}(0,\sigma))_i, \quad i = 1,...,N \tag{11}$$

where: Y_i is a new individual generated by crossover or mutation operation, X_i^1 and X_i^2 are parents individuals, $\mathcal{N}(mean,\sigma)$ denotes the normally distributed random variable, where $mean$ is a random number and σ its variation, respectively.

Moreover, in order to generate new population the classical roulette selection is used. The probability $\Pr(X)$ of receiving the individual X from population P, ($X \in P$) is:

$$\Pr(X) = \frac{fitness(X)}{\sum_{Y \in P} fitness(Y)}, \quad \forall X \in P \tag{12}$$

where: $fitness(X)$ is an estimation of the adaptation of the *X*–th individual (the value of goal function). The HGS-FP results for some test functions can be found elsewhere [4].

4. Results and Discussion.

To use the HGS strategy in diffusion process we should reformulate the problem into the optimization one (by finding the extreme). Let $N_i^{\exp}$, $i = 1,...,r$ be the molar fractions of the searching profile, i.e. the experimental results. The Hierarchical Genetic Strategy will minimize the metric sum:

$$d\left(N^{\exp}, N^{calc}\right) = \int_{-\Lambda}^{\Lambda} \omega \sum_{i=1}^{r} \left| N^{\exp} - N^{calc} \right| \tag{13}$$

where N^{calc} denote the calculated molar fractions (with the use of Eq. (1)) and ω is the weight function.

To show the efficiency of HGS-FP method, the intrinsic diffusion coefficients of Cu-Fe-Ni system at 1273 K were calculated. The Cu-Fe-Ni system was chosen because it is a single phase in a wide range of compositions and its thermodynamic properties are fairly well known [16].

The calculated concentrations profiles of Cu, Fe and Ni are compared with the experimental results [16]. The Darken method with the HGS-FP and Redlich-Kister approximation allows obtaining satisfactory agreement with experimental data, Fig. 1.

Fig. 2 shows that the inverse problem of interdiffusion is not unique (i.e. many global optima can be found).

The presented method of the quantitative description of the inverse method of chemical transport process in multicomponent alloy is self-consistent and is effective in practical computations. The simulation of interdiffusion in Fe-Ni-Cu diffusion couples shows satisfactory agreement with experimental data. Moreover the non-uniqueness of the inverse method to find the intrinsic diffusion coefficients was shown. To avoid such difficulties the fitness function should be changed, e.g.,. the time evolution of the experimental diffusion path should be known and included.

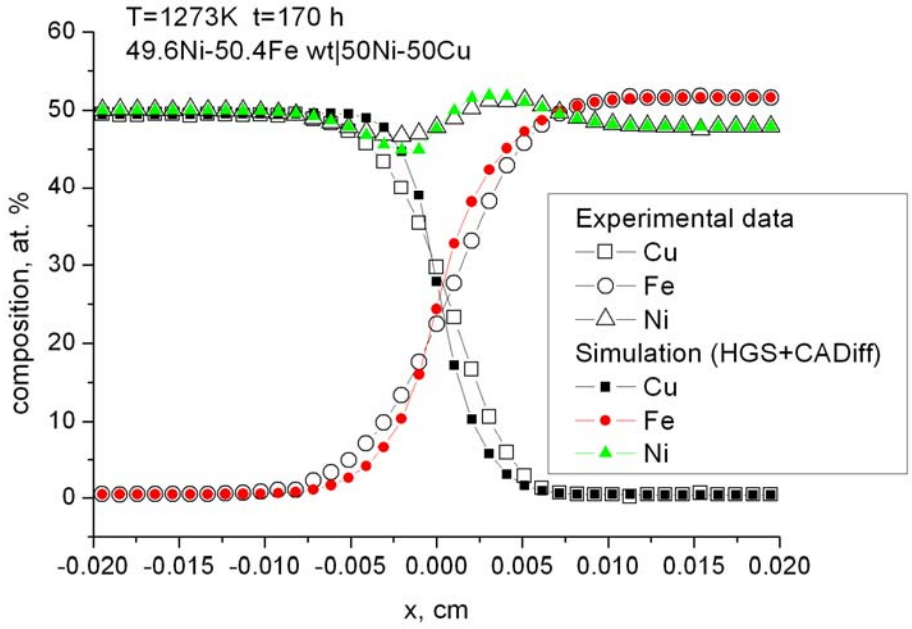

Fig. 1. Comparison of experimental and calculated diffusion profiles of Cu-Fe-Ni system at 1273 K.

The Hierarchical Genetic Strategy was searching for optimal intrinsic diffusion coefficients in the range of $10^{-13} - 10^{-9}$ [cm^2 s^{-1}]. The results of HGS-FP, local and global optima (i.e. the normalized intrinsic diffusion coefficients of Cu, Fe and Ni) are shown on Fig. 2.

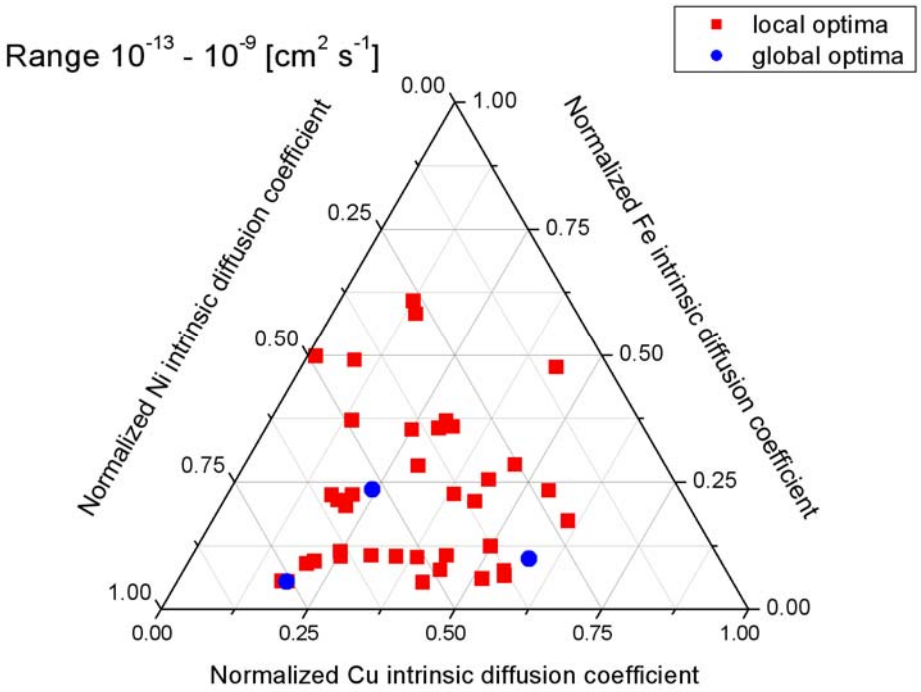

Fig. 2. The normalized diffusion coefficients in Cu-Fe-Ni diffusion couple at 1273 K.

Acknowledgements. MATERA ERA-NET EU project (MASTRA) is acknowledged for financial support.

1 Cantu-Paz E.; *Efficient and Accurate Parallel Genetic Algorithms*. Kluwer 2000.
2 Whitley D., Rana S., Hackendorn R.B; Island Model Genetic Algorithms and Linearly Separable Problems, AISB 1997 Workshop on Evolutionary Computing, Manchester.
3 Schaefer R., Kołodziej J., Gwizdała R., Wojtusiak J.; How simpletons can increase the community development - an attempt to hierarchical genetic computation. *Proc. of the 4th Conf. on Evolutionary Algorithms and Global Optimization*, Lądek Zdrój, 5-8 June 2000, pp. 187-198.
4 Wierzba B., Semczuk A., Kołodziej J., Schaefer R.; Hierarchical Genetic Strategy with real number encoding. *Proc. of the 6th Conf. on Evolutionary Algorithms and Global Optimization*, Łagów, 26-28 May 2003, pp. 231-237.
5 M.A. Dayananda, Y.H. Sohn, *Metall*. *Trans*. *A* 1999, 30 A, 535.
6 Darken L. S., Trans. A.I.M.E., 1948, 174, 184
7 Holly K., Danielewski M., Phys. Rev. B, 1994, 50, 13336
8 Nernst W., Z. Phys. Chem., 1889, 4, 129
9 Planck M., Ann. Phys. Chem., 1890, 40, 561
10 Danielewski M., Wierzba B.: *"Mechano-Chemistry; Diffusion in Multicomponent Compressible Mixtures",* Physica A 387 (2008) 745
11 Kaufman L. and Bernstein H.: „Computer Calculation of Phase Diagrams", Academic Press, New York 1970.
12 Dictra User's Guide, Version 21., www.thermocalc.com/Products/Dictra.html
13 Wierzba B., Semczuk A., Kołodziej J., Schaefer R.; Hierarchical Genetic Strategy with real number encoding. *Proc. of the 6th Conf. on Evolutionary Algorithms and Global Optimization*, Łagów, 26-28 May 2003, pp. 231-237.
14 Schaefer R., Sędziwy S.; Advances in multi-agent systems, Jagiellonian University, Cracow, Poland, 2001.
15 Kołodziej J., Schaefer R., Paszyńska A. : Hierarchical Genetic Computation in Optimal Design, J. of Theo. and App. Mech., Comp. Intell. 42, 2004, pp.519
16 . Bachorczyk-Nagy: "Metody wyznaczania współczynników dyfuzji i ewolucja składu w stopach wieloskładnikowych", PhD thesis, Kraków 2002 (in polish).

Одержано редакцією 2.12.2008
Прийнято до публікації 4.12.2008