



## CELLULAR AUTOMATA MULTI-COMPONENT DIFFUSION MODELS

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### Abstract

The paper presents a survey of developed multi-component diffusion models. Stochastic and continuous PDE models for multi-component diffusion problems are studied by performing a sequence of numerical simulations. The stochastic models are based on Cellular Automata with interchange and block-rotation model. The multi-component particles are either interchanged or rotated clockwise or counter-clockwise with prescribed probability. The diffusion coefficient is tuned by changing the probabilities of execution of particular rules. The continuous PDE models are based on classical variational formulations solved by the self-adaptive hp Finite Element Method. Numerical results are compared and discussed.

**Key words:** cellular automata, multi-component diffusion

### 1. INTRODUCTION

The Cellular Automata (CA) models for the multi-component diffusion presented in this paper are an extension of the single component diffusion models introduced by [1]. These models actually work in the time-space scale located between the macro and nano scales. It should be emphasized, that these models do not incorporate nano-scale interparticle interaction models nor the macro-scale continuous models, based on partial differential equations. The presented models utilize the assumption that the diffusion process can be simulated by random executing of some exchange or rotation rules, for grains of considered components. In other words,

these models do not express the physical laws directly at level of nano-scale particles of affected components, rather they define some simplified discrete rules at the level of some virtual grains of the components.

The trade-off is the necessity of the model calibration. The model is prescribed by means of the following parameters: the probability of execution of particular rules, and the number of rules that must be executed in order to simulate a given real time interval. According to [1], for a single component diffusion process, the more rules one executes in every simulation step, the faster the diffusion process and the higher the resulting diffusion coefficient will be.

Bandman [1] suggests maintaining the simulation until the concentration distribution will cover the data obtained from experiment or from other numerical method. The proportion of the number of executed rules to the time interval obtained from the experiment, or from other reliable simulation, allows one to find the correlation of the simulation step to the real time scale. E.g., if the number of rules  $N_{ORG}$  executed within a single simulation step results in  $D_{ORG}$  diffusion coefficient (which has been verified by comparison either with experiment or with other simulation results), the other diffusion coefficient  $D_{NEW}$  can be obtained with the following number of rules (executed with a single simulation step):

$$N_{NEW} = N_{ORG} \frac{D_{ORG}}{D_{NEW}} \quad (1)$$

It is also possible to tune the diffusion coefficient by changing probabilities of execution of particular rules [1]. In this paper, the models discussed by [1] are extended to the multi-component situation. The correspondence between the number of rules executed within the simulation step with the real time scale are found by comparison of numerical experiments with data obtained from other reliable 1D simulation [2-5]. Thus, in every simulation step, the number of particles and the transformation rules to be executed are selected randomly. The crucial assumption that was made for the multi-component simulation is that in every simulation step in the total number of selected particles there are  $N_m$  particles of  $m^{\text{th}}$  component, and these numbers are determined by the proportion of the known diffusion coefficients of particular components. In other words, the proportion of the transformed particles of different components is kept equal to the proportion of the diffusion coefficients.

## 2. DIFFUSION PROBLEM

In this section a review of the existing numerical models for single component diffusion problem is presented. The CA models are introduced by using new formalism, based on the assumption that CA simulation can be interpreted as a sequence of matrix transformations.

### a. Partial Differential Equation (PDE) strong formulation

The classical diffusion problem can be formulated by the second Fick's law, i.e. find

$R^n \times [t_o, t_N] \ni \Omega \times I \ni (x, t) \rightarrow c(x, t) \in R$  the concentration  $c$  distribution scalar field such that

$$\frac{\partial c}{\partial t} - \nabla \cdot (D \nabla c) = 0 \quad \text{on } \Omega \times I \quad (2)$$

where  $D$  stands for the diffusion coefficient. The diffusion coefficient can be defined as a function of concentration

$$R^n \times [t_o, t_N] \ni \Omega \times I \ni (x, t) \rightarrow c(x, t) \rightarrow D(c) \in R \quad (3)$$

prescribed in the domain  $\Omega$ , possibly changing in time. The solution of the problem inside the area  $\Omega$  is prescribed by the initial and boundary conditions. The Dirichlet boundary condition

$$c = c_D \quad \text{on } \Gamma_D \times I \quad (4)$$

defines the concentration field on the  $\Gamma_D$  part of the boundary, while the Neumann b.c.

$$\mathbf{n} \cdot \nabla c = g \quad \text{on } \Gamma_N \times I \quad (5)$$

defines the flux  $g$  on  $\Gamma_N$  boundary. The initial condition defines the starting configuration of the concentration field

$$c = c_0 \quad \text{on } \Omega \times I \quad (6)$$

Thus, in the simplified model described here, the concentration inside the domain  $\Omega$  depends only on the concentration and the flux on the boundary, and given initial configuration of the concentration field (i.e. the chemical potential gradients and other forces are neglected). The configuration of the initial and boundary conditions depend on the problem being solved, thus the considerations are not illustrated with any figure.

### b. Weak (variational) formulation

The weak (variational) formulation is obtained by considering  $L_2$  scalar products with test functions from  $V = H^1(\Omega)$  space [6-7]. The weak problem can be formulated in the following way: Find the concentration distribution  $c \in c_D + V$  scalar fields satisfying

$$(\dot{c}, v)_\Omega + \int_\Omega D \nabla c \circ \nabla v d\Omega = 0 \quad \forall v \in V \quad (7)$$

$$(c(0), v)_\Omega = (c_0, v)_\Omega \quad \forall v \in V \quad (8)$$

The necessary discretization in time implies the following matrix system for both problems

$$\mathbf{M}\dot{\mathbf{u}} + \mathbf{K}\mathbf{u} = \mathbf{f} \quad (9)$$



Applying the trapezoidal rule with respect to the time derivative [6-7] we obtain

$$(\mathbf{M} + \alpha\delta\mathbf{K})\mathbf{u}^{k+1} = [\mathbf{M} - (1 - \alpha)\delta\mathbf{K}]\mathbf{u}^k + \delta\mathbf{f}^k \quad (10)$$

where  $\mathbf{M}$  is the mass matrix,  $\delta$  is the time step,  $\alpha \in [0,1]$  gives different time integration schemes.

**c. General cellular automata formulation**

The diffusion problem can be also solved by means of the Cellular Automata (CA). The CA model consists of a matrix  $U(N \times N)$  of  $N \times N$  finite automata (cells) interacting with close neighbours.

**Definition 1.** The *state* of each cell  $\{u[i, j]\}_{i,j=1,\dots,N}$  is prescribed by the state function

$$(i, j) \rightarrow u[i, j] \in R^n, \quad i, j = 1, \dots, N \quad (11)$$

where  $n$  is the number of state variables.

**Definition 2.** The behaviour of each cell is prescribed by several *transition rules*  $\theta_{i,j}^k : U(N \times N) \rightarrow U(N \times N)$ . Each transition rule can be formally defined as the sum of several matrix transformations  $\theta_{i,j}^{((a,b),(c,d))} : U(N \times N) \rightarrow U(N \times N)$  as

$$\theta_{i,j}^k = \bigcup_{((a,b),(c,d)) \in S_{i,j}^k} \theta_{i,j}^{((a,b),(c,d))} \quad (12)$$

where  $S_{i,j}^k$  is a set of cells affected during the execution of the  $k^{\text{th}}$  transition rule for the cell  $(i, j)$ . Each matrix transformation  $\theta_{i,j}^{((a,b),(c,d))}$  actually replaces terms  $u[a, b]$  and  $u[c, d]$  in the matrix  $U$ . The transformation is formally defined as the following

$$\theta_{i,j}^{((a,b),(c,d))}U = U - G_aUG_b - G_cUG_d + P_{a,c}(G_aUG_b + G_cUG_d)P_{b,d} \quad (13)$$

where  $P_{i,j}$  is the permutation matrix, e.g. the  $P_{2,4}$  permutation matrix for  $U(5 \times 5)$ , defined as

$$P_{2,4} = \begin{bmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{bmatrix}, \quad (14)$$

replaces the second and the fourth rows if it is applied from the left, while it replaces the second and the fourth column if it is applied from the right. The

$G_i$  matrix is the zero matrix with 1 at the diagonal in  $i^{\text{th}}$  row, e.g.  $G_2$  defined as

$$G_2 = \begin{bmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix} \quad (15)$$

sets the entire matrix to 0 except the second row if it is applied from the left, and sets the entire matrix to 0 except the second column if it is applied from the right. Summing up, the equation (13) is just a formal definition of replacement of  $u[a, b]$  and  $u[c, d]$  terms in the matrix  $U$ . Thus, for each cell  $(i, j)$  there are  $k$  transition rules defined. The probability of execution of each rule is globally prescribed by probabilities

$$P(\theta) = (p_1, \dots, p_k) \quad (16)$$

**Definition 3.** The *CA simulation* consists in execution of sequence of transformations

$$\{\theta_{i,j}^k\}_{(i,j,k) \in H} \quad (17)$$

where  $H$  is a history of the CA simulation.

**Algorithm 1.** The classical CA simulation algorithm can be summarized as follows:

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Loop wrt. the number of simulation steps
  Loop wrt. the number of performed transformations M
    Select randomly a cell (i, j) with uniform probability
    Select randomly transition rule k with probabilities P(theta)
    Execute the transition rule theta_{i,j}^k over the cell (i, j)
    
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Notice, that the transition rule is defined by providing the set of interchanged particles  $S_{i,j}^k$  from (11), and the transition applicability probabilities  $P(\theta) = (p_1, \dots, p_k)$  from (16). Following [1], the CA algorithm can be expressed either by the interchange or block-rotation rules.

**d. Interchange CA formulation**

In the interchange CA formulation [8], the transition rules are defined as follows:



$$P(\theta) = \left( \frac{1}{4}, \frac{1}{4}, \frac{1}{4}, \frac{1}{4} \right) \quad (18)$$

$$S_{i,j}^1 = \{((i,j),(i-1,j))\} \quad S_{i,j}^2 = \{((i,j),(i+1,j))\}$$

$$S_{i,j}^3 = \{((i,j),(i,j-1))\} \quad S_{i,j}^4 = \{((i,j),(i,j+1))\} \quad (19)$$

In this model, there are four different rules, responsible for interchange of particle  $(i,j)$  with one of its four neighbours, selected randomly. According to [1], the diffusion coefficient of this model can be modified by adjusting the number of state transitions executed in each simulation step of the simulation.

**e. Block-rotation CA model**

In the block-rotation CA formulation [9], the transition rules are defined as follows:

$$P(\theta) = \left( \frac{1}{2}, \frac{1}{2} \right) \quad (20)$$

$$S_{i,j}^1 = \{((i,j),(i,j+1)),((i,j+1),(i+1,j+1)),((i+1,j+1),(i+1,j)),((i+1,j),(i,j))\}$$

$$S_{i,j}^2 = \{((i,j),(i+1,j)),((i+1,j),(i+1,j+1)),((i+1,j+1),(i,j+1)),((i,j+1),(i,j))\}$$

$$(21)$$

In this model particle  $(i,j)$  is either clock-wise or counter clock-wise rotated with its four neighbours. According to [1], the diffusion coefficient of this model can be modified either by adjusting the number of state transitions executed in each simulation step or by modifying the probability coefficients (20).

**3. MULTI-COMPONENT DIFFUSION PROBLEM**

In this section, the extension of the single component diffusion model to the multi-component one is discussed. The continuous description, be means of the strong (2)-(6) or weak (7)-(8) equations, has the following limitations. In the case of the multi-component simulation, the diffusion function for the model depends on the local concentration, which makes the problem non-linear. This implies a complex numerical simulation.

The CA models are free from the above limitations, since we can distinguish different states of particles representing different components, and can modify the transition rules accordingly. For  $d$  components diffusion problem, the state function is defined as follows

$$(i,j) \rightarrow u[i,j] \in \{u_1, u_2, \dots, u_d\} \quad i, j = 1, \dots, N. \quad (22)$$

To reflect different diffusion coefficients of each component, the following modification to the general CA simulation algorithm is made.

**Algorithm 2.** The multi component CA simulation algorithm utilizing the vector of prescribed diffusion coefficients for all components  $[D_1, D_2, \dots, D_d]$  is defined as follows:

Loop wrt. the number of simulation steps

    Loop wrt. the number of performed transformations  $M$

        Select randomly a component type (denoted by  $m$ )

            with the probability  $N_m$

            Select randomly a cell  $(i,j)$  of type  $m$

            Select randomly transition rule  $k$  with  $P(\theta)$

            Execute the transition rule  $\theta_{i,j}^k$  over the cell  $(i,j)$

        where

$$N_m = \left( \frac{D_m}{\sum_{k=1, \dots, d} D_k} \right) \quad (23)$$

The proportion between intrinsic diffusion coefficient of different components determines the transformation probability of particles of a particular kind at each simulation step. The presented modification can be used for either the interchange or block-rotation models.

**4. REFERENCE 1D MODEL**

For verification of our 2D multi-component CA algorithm, we have refered to the 1D model implemented within the CADiff software [2-3]. The CADiff method is based on the mass conservation law:

$$\frac{\partial c_i}{\partial t} = -\nabla \circ J_i^d - \nabla \circ J_i^D \quad i = 1, \dots, r \quad (24)$$

where:  $c_i$  denote the concentration of the  $i$ -th component,  $J_i^d$  and  $J_i^D$  denotes the diffusion and darken flux of  $i$ -th component, respectively. The overall flux is defined as a sum of the diffusion and Darken fluxes,  $J_i = J_i^d + J_i^D$ . The Darken flux,  $J_i^D$ , is generated during interdiffusion due to locally unbalanced diffusion fluxes [4]. We used here the simplifying assumption of constant total molar concentration in the alloy:



$$J_i^D = c_i v^D = -c_i \sum_{j=1}^r \frac{c_j}{c} v_j^d \quad (25)$$

where:  $v^D$  - Darken velocity that is common for every component,  $v_i^d$  - the diffusion velocity of the  $i$ -th component,  $c = \sum_{i=1}^r c_i = const.$  - the overall molar concentration in the alloy.

**Table 1.** Composition of Fe-Ni-Cr and other components in SS304|SS310 diffusional couple, according to [6].

Alloy	Fe	Ni	Cr	Other
SS 304	70%	10%	18%	2%
SS 310	52%	22%	25%	1%

**Table 2.** Intrinsic diffusion of Cr, Fe and Ni at 1373 K [6, 7].

	Fe	Ni	Cr
Intrinsic diffusion coefficient $D_i$	$D_{Fe} = 1.92 \cdot 10^{-15}$	$D_{Ni} = 1.10 \cdot 10^{-15}$	$D_{Cr} = 2.79 \cdot 10^{-15}$

**Table 3.** Probability distribution of transformation of particles of particular components (Cr, Fe and Ni) in one simulation step.

	Fe	Ni	Cr
$N_m$	192/581	110/581	279/581

The diffusion flux as in previous section was defined by Fick's law:

$$J_i^d = -D_i \nabla c_i \quad i = 1, \dots, r \quad (26)$$

### 5. NUMERICAL EXPERIMENTS

The numerical experiment concerns a two dimensional simulation of the diffusion process between two alloys: SS 304 and SS 310 with respective compositions, presented in table 1. The two alloys have been put into contact and heated up to 1373 K. The intrinsic diffusion coefficients of all components in the temperature of 1373 K are presented in table 2. The simulations have been performed in 2D with the interchange and block rotation CA models, with the multi-component simulation Algorithm 2.

The multi-component simulation algorithm in every simulation step performs a transformation of particles of the Fe element with the probability  $N_1$ , a transformation of particles of the Ni element with the probability  $N_2$  and a transformation of particles

of the Cr element with the probability  $N_3$ . The value of  $N_m$  is obtained from formula (23).

In other words, within one simulation step, the particles of the Fe element are transformed with the probability 192/581, the particles of the Ni element are transformed with the probability 110/581 and the particles of the Cr element are transformed with the probability 279/581. These are summarized in table 3.

The simulation results are presented in figure 1. The 1D plot of the concentration of all components during the simulation has been compared to the distributions obtained from other 1D numerical simulation [2], based on the experimental data [10]. The verification of our 2D model is based on the estimation of the concentrations  $c_m(x)$  of the three components (Fe – Ni – Cr) over the vertical cross-sections of the 2D domain, made according to the following rule:

$$c_m(x) = \frac{K_m(x)}{K(x)} \quad (27)$$

where  $K(x)$  is the number of grains (cells) over the vertical cross-section at horizontal coordinate  $x$ ,  $K_m(x)$  is the number of grains (cells) of  $m^{\text{th}}$  component over the vertical cross-section at horizontal coordinate  $x$ . The data obtained from the experiment [10] are presented on the last panel in figure 1. There are two additional elements Al and Ir, whose contributions have been neglected in our numerical simulations. The 1D simulation results [5] are presented on the left panels in figures 2 and 3. The results of the CA simulation with Algorithm 2 for the interchange model are presented on the right panel in figure 2.

The results of the CA simulation with Algorithm 2 for the block-rotation model are presented on the right panel in figure 3. According to [1], the number of the simulation steps corresponding to the prescribed time interval can be obtained by comparing with experimental data or other reliable numerical simulations. In this case, the simulation results were compared with the 1D simulation from [5]. The simulation was stopped when the best match had been achieved, which made it possible to find the proportion of the number of transformation rules executed within one simulation step to the diffusion coefficients prescribed in the 1D simulation from [5]. The best match resulted in the difference between the concentration field of the order of 1%, with local jumps up to 4-5%, see figure 4.





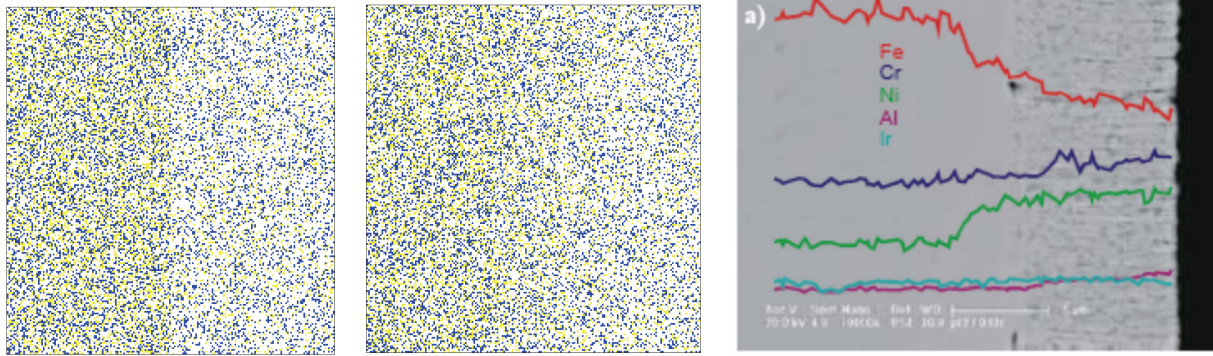


Fig. 1. The initial and final configurations of cells, and the cross-section of two alloys obtained from the experiment [7], with plots denoting concentration of particular elements.

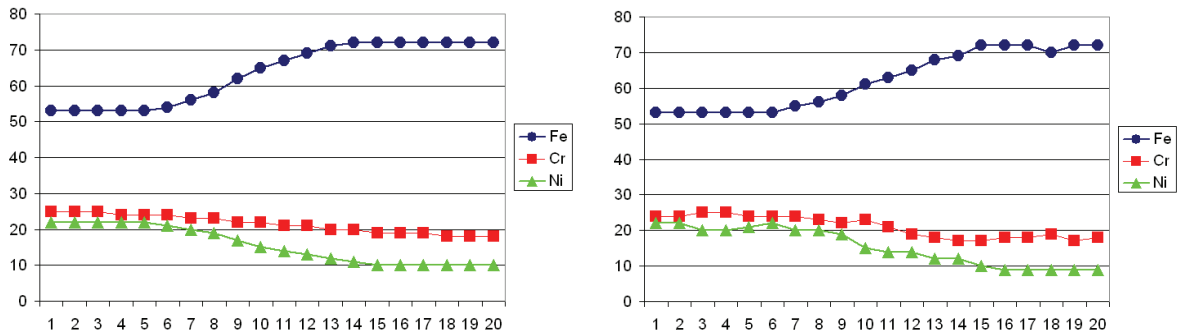


Fig. 2. Comparison of the 1D simulation [2] with interchange CA model simulation.

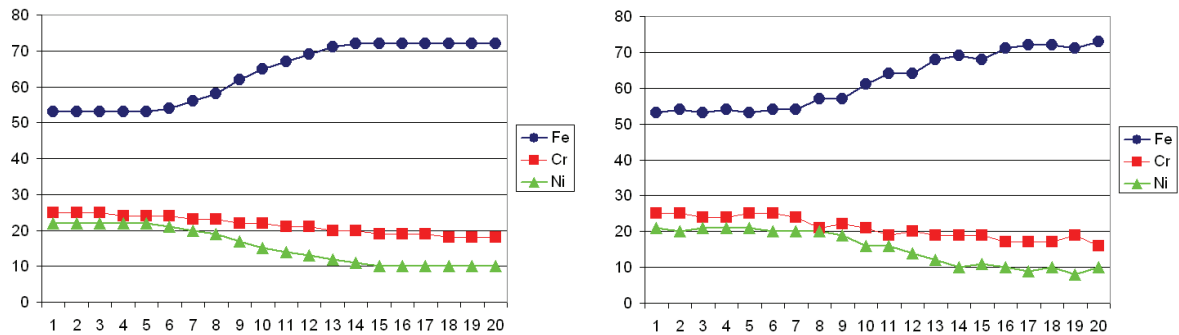


Fig. 3. Comparison of the 1D simulation [2] with block-rotation CA model simulation.

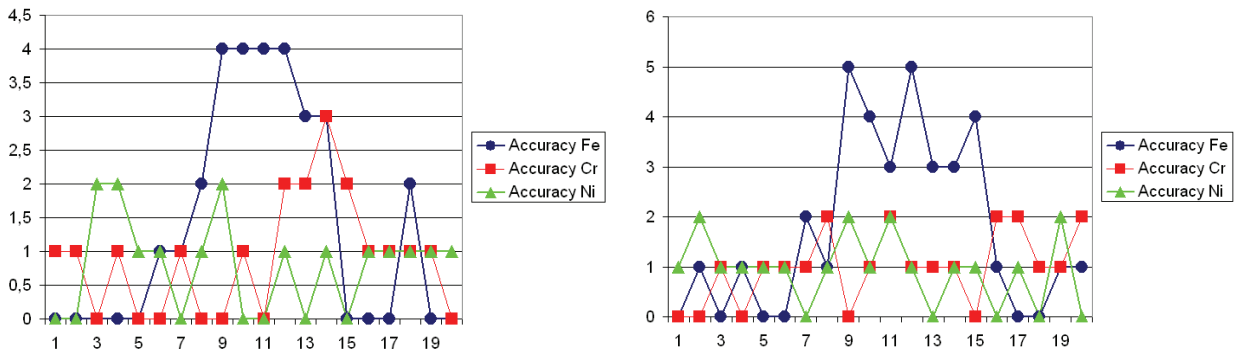


Fig. 4. % error of the interchange and block-rotation CA model simulations.



## 6. CONCLUSIONS

An extension of the CA models presented by [1] to the multi-component diffusion simulations has been presented. The models utilize an analogy of the diffusion process with a simple phenomenon of interchange or block rotation of adjacent grains of different components. It is postulated that such a simple model can be successfully utilized to simulate complex physical phenomena of the multi-component diffusion. This can be done if the model parameters, such as the probabilities of selection of grains of different components and the number of grains transformed during one simulation step, are selected in a proper way. It has been showed that the probabilities of selection of particles of different components for the transformation within a single simulation step must be defined according to the proportion of the intrinsic diffusion coefficients of the three components. The number of the transformations has been selected by comparison with other 1D numerical simulations [2-5] based on the experimental data [10]. The obtained results are satisfactory. It is postulated that the model can be easily utilized for more components as well as in fully 3D simulations.

## ACKNOWLEDGEMENT

The work has been financially supported by Ministry of Scientific Research and Information Technology grant no. 3 TO8B 055 29.

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## MODEL AUTOMATÓW KOMÓRKOWYCH DYFUZJI WIELOSKŁADNIKOWEJ

Streszczenie

W artykule zaprezentowany został przegląd opracowanych modeli dyfuzji wieloskładnikowej. Problem dyfuzji wieloskładnikowej opisany został z pomocą modeli stochastycznych, bazujących na automatach komórkowych, jak również modeli ciągłych, bazujących na równaniach różniczkowych cząstkowych.

Rozważane modele stochastyczne opierają się na automatach komórkowych z regułami wymiany lub rotacji cząstek wielofazowych. Podczas symulacji, cząstki różnych faz poddawane są rotacji zgodnie lub przeciwnie do ruchu wskazówek zegara, zgodnie z przyjętym prawdopodobieństwem. Współczynnik dyfuzji symulowanego układu cząstek definiowany jest poprzez modyfikację prawdopodobieństw wykonania reguł międzycząsteczkowych. Opracowane modele zostały zweryfikowane poprzez wykonanie szeregu symulacji numerycznych, oraz porównane do uprzednio opracowanych modeli ciągłych, bazujących na adaptacyjnej metodzie elementów skończonych.

*Submitted: October 8, 2008*

*Submitted in a revised form: November 7, 2008*

*Accepted: November 20, 2008*

