Available online at www.sciencedirect.com

ScienceDirect

Ceramics International 36 (2010) 1983–1988

CERAMICS
INTERNATIONAL

www.elsevier.com/locate/ceramint

Monte Carlo simulation of microstructure evolution in biphasic-systems

Devis Bellucci *, Valeria Cannillo, Antonella Sola

Dipartimento di Ingegneria dei Materiali e dell'Ambiente, Università degli Studi di Modena e Reggio Emilia, Via Vignolese 905, 41100 Modena, Italy

Received 23 February 2010; received in revised form 30 March 2010; accepted 20 April 2010
Available online 11 June 2010

Abstract

Over the past few decades, a variety of models have been proposed in order to investigate the grain growth kinetics and the development of crystallographic textures in polycrystalline materials. In particular, a full understanding of the microstructure evolution is a key issue for ceramic systems, since their mechanical or thermal behaviour is intimately related to their microstructure. Moreover, the development of appropriate simulative tools is crucial to reproduce, control and finally optimize the solid-state sintering process of ceramics. Monte Carlo simulations are particularly attractive because of their ability to reproduce the statistical behaviour of atoms and grain boundaries with time. However, Monte Carlo simulations applied to two-phase materials, such as many ceramic systems, result complex because both grain growth and diffusion processes should be taken into account. Here the Monte Carlo Potts model, which is widely used to investigate the crystallization kinetics for monophasic systems, is modified and extended to biphasic ones. The proposed model maps the microstructure onto a discrete lattice. Each lattice element contains a number representing its phase and its crystallographic orientation. The grain formation and growth are simulated by appropriate switching and reorientation attempts involving the lattice elements. The effect of temperature is also discussed.

Keywords: A. Grain growth; B. Grain boundaries; B. Grain size; Monte Carlo simulations

1. Introduction

Computer simulation of microstructure evolution is critical in material science. In fact, many materials properties such as viscosity, brittleness, etc., are strictly connected to their crystalline texture. Among microstructural phenomena, the description and modelling of grain growth kinetics in polycrystalline materials is of primary interest for metallurgy and ceramic technology [1], in particular to optimize ceramics sintering processes according to prescribed conditions. Thanks to the increase in computer processing power, in the last 20 years an accurate computer simulation of grain growth has become possible.

In the mid-'80s, Anderson and co-workers [2–5] proposed a new method to simulate grain growth. This approach, which is currently the most frequently used for these simulations, combines the Monte Carlo procedure (MC) with the Potts model [6]. Originally the Potts model was developed as an extension to the ferromagnetic Ising model for systems with arbitrary spin degeneracy. In the Ising model [7], which is used to model simple magnets, every lattice site of a two-dimensional array is assigned a magnetic spin state, i.e. spin up or spin down. The energy of the system is determined by the sum of the interactions between a spin and its neighbours, and the system itself evolves minimizing the boundary between domains with the same spin. In the Potts model, the twofold degeneracy of the Ising systems was extended to an arbitrary number Q of magnetic spin states, giving rise to domain structures quite similar to the grain structures in polycrystalline materials. In fact, both systems have infinite degeneracy, since in a certain volume of material there can be a large number of crystallographic orientations. In addiction, both systems deal with a two- or three-dimensional array of interacting elements that evolve in order to minimize the boundary area.

The Potts model was firstly applied to investigate the microstructural evolution and grain growth in isotropic single phase systems [2]. An increase of the average grain size was observed with time, leading to the formation of large grains at the expense of small grains. Moreover, it was demonstrated that the average grain radius $\langle R \rangle$ increases with time t according to

$$\langle R \rangle \approx t^n \quad (1)$$

---

* Corresponding author. Tel.: +39 059 2056281; fax: +39 059 2056243.
E-mail address: devis.bellucci@unimore.it (D. Bellucci).

0272-8842/$36.00 © 2010 Elsevier Ltd and Techna Group S.r.l. All rights reserved.
doi:10.1016/j.ceramint.2010.05.006

where $n = 0.5$ in both two- and three-dimensional systems [8]. Eq. (1) confirmed the previsions of most of the grain growth theories [9,10] and was the first major success of the Potts model applied to materials science. However, Eq. (1) refers only to an average growth. As observed by Anderson et al. [2], although large grains generally grow at the expenses of small ones that shrink, it is possible to find instances where the opposite behaviour is true. Therefore, it is complex to predict the evolution of individual grains.

After the abovementioned pioneering work, the Potts model coupled with MC procedure has been employed to evaluate a variety of materials systems, generating snapshots of the evolving microstructure with time. In fact, one of the main advantages of these techniques is the possibility to visualize graphically, even dynamically, the grain growth process. To this aim, snapshots of the evolving system with time are generated and compared to the real microstructures, often with the help of image analysis software. Among the microstructural phenomena simulated, the study of particle pinning effect on grain boundary [11], recrystallization [12], abnormal grain growth [13], crystal growth from the melt (solidification) [14], grain growth by Ostwald ripening [15], and the investigation of the boundary character during annealing can be mentioned [17]. An overview on the MC Potts model for simulating microstructural evolution can be found in [18].

Although many works in the literature deal with MC Potts simulations for single phase polycrystalline materials [11–19], this technique has been rarely applied to biphasic-systems. In this case the crystallization kinetics is more complex, because both grain growth and diffusion of the two phases components within the system should be taken into account.

In this work, the MC Potts model is modified in order to study two-phase systems. As reported in the next section, during the diffusion process, for a given element of the system a wider range of interactions than the six-nearest-neighbours one is taken into account, which makes it possible to describe both grain boundary and phase migration. The effect of temperature, i.e. of thermal "noise" introduced into the lattice representing the system, is also discussed.

2. Description of the numerical method

As already mentioned, the crystallization process occurs by means of grain boundary migration, which determines the shrinkage of some grains and the contemporaneous growth of other grains. The most important driving force for grain growth is the reduction of the excess surface free energy associated to the total grain boundary area of the system. At the end, the system is expected to reach a steady state where the normalized grain size distribution becomes invariant with time.

The model proposed in this work maps the material and its grain microstructure onto a discrete 2D lattice with full periodic boundary conditions. Each site corresponds to a surface element and contains a number whose sign ("+" or "−") denotes which phase the element belongs to, i.e. positive numbers represent phase "a" elements, while negative numbers refer to phase "b". The absolute value of the number, ranging between 1 and C, represents the element crystallographic orientation. Within the same phase, a grain is defined by a cluster of elements containing the same number. The grain boundaries divide two grains of unlike crystallographic orientation or dissimilar phase. A 2D matrix exemplifying the material and its grain structure is reported in Fig. 1. In particular, in this picture a graphic solution has been adopted to plot the matrix so that the triangular shape of the lattice is visible. Fig. 1 evidences the presence of two different phases as well as the grain structure within each phase. A brief discussion on the lattice effects in Potts model simulations is reported in the next section.

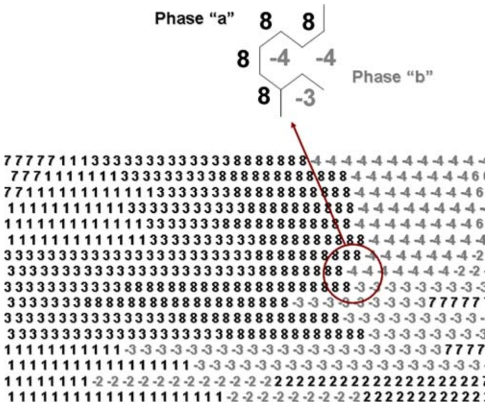


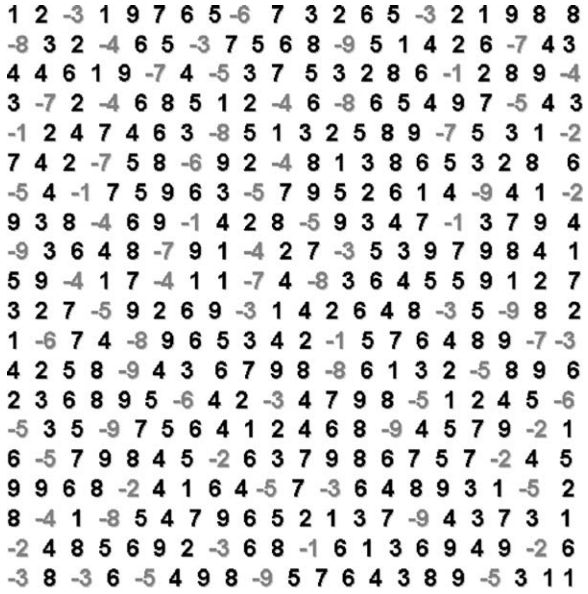
Fig. 1. The grain texture bitmapped onto a triangular lattice. Positive black numbers stand for phase "a" elements, while negative grey numbers stand for phase "b".

The starting distribution of orientations C is chosen at random. Also positive and negative elements are randomly distributed, but their amount is fixed according to the volume fractions of the respective constituent phases. Fig. 2 represents, as an example, a starting matrix where each element i is assigned a sign and a random crystallographic orientation C_i between 1 and C.

Then, the system evolves in order to reduce the number of nearest-neighbour pairs of unlike crystallographic orientation or belonging to different phases. Thanks to the introduction of specific energy parameters, the developed algorithm accounts for the different interactions that may occur between elements belonging to the same phase or between elements belonging to two distinct phases. The main steps of the algorithm can be summarized as follows:

(a) Random choice of an element J of the matrix.
(b) Calculation of the initial free energy G_{in} of the element J with its current crystallographic orientation C_j, based on its six-nearest-neighbour elements. The thermodynamic of atomic interactions between the considered element J and a given nearest-neighbour element differs depending on whether they belong to the same phase or not.

The Hamiltonian that describes the energy G of a lattice element surrounded by six nearest neighbours belonging to the



1	2	-3	1	9	7	6	5	-6	7	3	2	6	5	-3	2	1	9	8	8
-8	3	2	-4	6	5	-3	7	5	6	8	-9	5	1	4	2	6	-7	4	3
4	4	6	1	9	-7	4	-5	3	7	5	3	2	8	6	-1	2	8	9	-4
3	-7	2	-4	6	8	5	1	2	-4	6	-8	6	5	4	9	7	-5	4	3
-1	2	4	7	4	6	3	-8	5	1	3	2	5	8	9	-7	5	3	1	-2
7	4	2	-7	5	8	-6	9	2	-4	8	1	3	8	6	5	3	2	8	6
-5	4	-1	7	5	9	6	3	-5	7	9	5	2	6	1	4	-9	4	1	-2
9	3	8	-4	6	9	-1	4	2	8	-5	9	3	4	7	-1	3	7	9	4
-9	3	6	4	8	-7	9	1	-4	2	7	-3	5	3	9	7	9	8	4	1
5	9	-4	1	7	-4	1	1	-7	4	-8	3	6	4	5	5	9	1	2	7
3	2	7	-5	9	2	6	9	-3	1	4	2	6	4	8	-3	5	-9	8	2
1	-6	7	4	-8	9	6	5	3	4	2	-1	5	7	6	4	8	9	-7	-3
4	2	5	8	-9	4	3	6	7	9	8	-8	6	1	3	2	-5	8	9	6
2	3	6	8	9	5	-6	4	2	-3	4	7	9	8	-5	1	2	4	5	-6
-5	3	5	-9	7	5	6	4	1	2	4	6	8	-9	4	5	7	9	-2	1
6	-5	7	9	8	4	5	-2	6	3	7	9	8	6	7	5	7	-2	4	5
9	9	6	8	-2	4	1	6	4	-5	7	-3	6	4	8	9	3	1	-5	2
8	-4	1	-8	5	4	7	9	6	5	2	1	3	7	-9	4	3	7	3	1
-2	4	8	5	6	9	2	-3	6	8	-1	6	1	3	6	9	4	9	-2	6
-3	8	-3	6	-5	4	9	8	-9	5	7	6	4	3	8	9	-5	3	1	1

Fig. 2. A typical initial 20 × 20 2D matrix, which simulates a system containing 80% phase "a" (positive black numbers) and 20% phase "b" (negative grey numbers).

same phase is

$$H = -V \sum_{nn} (\delta_{C_j C_k} - 1) \tag{2}$$

where C_j is the orientation of the lattice point J whose orientation change is being attempted, C_k is the orientation of a K nearest neighbour, V is the interaction energy between elements belonging to the same phase (expressed in units of the Boltzmann constant k_B), $\delta_{C_j C_k}$ is the Kronecker δ function, which is 1 if $C_j = C_k$ (sites with same orientation) and 0 otherwise (sites with different orientation). The summation is taken over all the six nearest neighbours of site J (i.e. $K = 1, \ldots, 6$). Therefore, neighbours belonging to the same phase contribute 0 to the system energy if they have the same orientation and V if not. Instead, the interaction between neighbouring elements belonging to two distinct phases results in a higher energy contribution, which is assumed to be equal to $6V$ regardless of their crystal orientations. Fig. 3 reports several examples of free energy calculation.

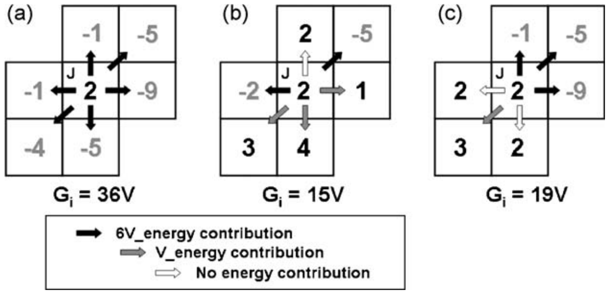


Fig. 3. Examples of calculation of the element J free energy with the considered closest neighbours.

(c) Random choice of an element Y among the nearest neighbours of J. From now on, there are two possibilities:

1. J and Y belong to the same phase. In this case a grain growth step is simulated by changing the crystallographic orientation of the element J. The algorithm can be schematized as follows:
   - (a) Random choice of a new crystallographic orientation C_f for the element J.
   - (b) Calculation of the free energy G_{fin} of the element J with its new crystallographic orientation C_f.
   - (c) Evaluation of $\Delta G = G_{\text{fin}} - G_{\text{in}}$, i.e. the change in system energy associated with the reorientation of the element lattice J. If the new orientation minimizes the energy, it is chosen. If the energy increases, the new orientation is not refused *a priori*, but it is accepted with a probability Π that depends on the system temperature.
2. J and Y belong to two distinct phases. In this case the algorithm simulates a transport step by switching the elements J and Y and, at the same time, by changing their crystallographic orientation.

(a) Calculation of the free energy G^y_{in} for the element Y with its present crystallographic orientation.

(b) The elements J and Y are interchanged.

(c) Random choice of a new crystallographic orientation C_f for J and C^y_f for Y (in order to preserve the relative amounts of the constituent phases, the orientation is tentatively changed, but not the sign, ±).

(d) Calculation of the free energy G_{fin} for J and G^y_{in} for Y. By means of this step and the next one, a wider range of interactions is taken into account during the switching process.

(e) Evaluation of $\Delta G = (G_{\text{fin}} + G^y_{\text{fin}}) - (G_{\text{in}} + G^y_{\text{in}})$. If the switching between J and Y and the associated reorientation of the two lattice elements minimize the energy of the system, the new positions and the new orientations are chosen. Otherwise, also in this case, the change is not rejected *a priori*, but it is accepted with a probability Π that depends on the system temperature. Fig. 4 reports a diagram of the described steps.

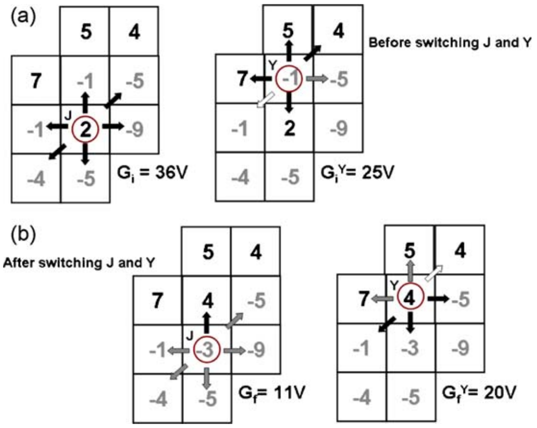


Fig. 4. Examples of calculation of the free energy G for the elements J and Y, before and after their switching. The switching minimizes the free energy of the system.

The reorientation (or the switching and reorientation) for a given step is performed with probability $\prod$:

$$\Pi = \begin{cases} \exp\left(\frac{-\Delta G}{k_B T}\right) & \Delta G > 0; \\ 1 & \Delta G \leq 0; \end{cases} \tag{3}$$

where ΔG is the variation in free energy calculated as previously described, T is the temperature and k_B is the Boltzmann constant.

The temperature T in Eq. (3) is not directly related to physical temperature, but it is a correction factor introduced to take care of specific lattice effects. T alters the transition probability Π and allows noise to be introduced in to the lattice.

For $T = 0$ (''static'' lattice), only micro-trials that minimize the free energy G are chosen. In case of $T \neq 0$ (''dynamic'' lattice), the temperature is another impelling force, together with the free energy lowering, that drives the physics of the system. The algorithm calculates Π and chooses a random number ρ between zero and one. If $\Pi > \rho$ then the micro-trial is chosen, even if $\Delta G > 0$. When $T \to \infty$, $\Pi \to 1$ and the system never crystallizes.

This sequence of steps, which is a Monte Carlo micro-trial, is repeated millions of time in order to simulate the free energy lowering of the system. The number of reorientation attempts is related to the simulation time through a quantity called Monte Carlo step (MCS). The MCS is equal to 1 when the number of simulation attempts (i.e. the number of Monte Carlo micro-trials) is equal to the number of elements in the lattice.

The simulation worked on 300×300 square matrixes. The maximum number of possible orientations C for each phase was set to 150. A triangular lattice was used to map the system, which means that each site has six nearest neighbours to be considered for energy calculations, coherently with the procedure previously described.

In order to test the biphasic algorithm, various composite systems, with different fractions of the constituent phases, were considered and their evolutions at different temperatures and times were simulated.

3. Results and discussion

As previously mentioned, in the present contribution 150 orientations were considered for each phase. Radhakrishnan et al. proposed N^2, where N is the dimension of the matrix [20]. For example, in a 300×300 matrix, $C = 300^2 = 90.000$. However, Srolovitz and coworkers [2] performed a calculation for a single phase system, observing that for $C > 30$ the simulation becomes C-independent. Taking into account these considerations, Blikstein and Tschiptschin [19] performed a Monte Carlo simulation of grain growth with $C = 128$ and a 200×200 square matrix. In particular, they observed that much higher C values, up to 30,000, did not improve the results, which remained analogous to the ones with $C > 64$.

As regards the lattice geometry the system is mapped on, many works in the literature are based on square [19] (with four nearest neighbours) and triangular lattices (with six nearest neighbours) [21]. The main goal is to reduce spurious lattice effects, which can alter the simulation and lead to non-physical results. As reported by Kim et al. [22], square lattices can lead to grain shapes that mimic the lattice symmetry, in fact the grain boundaries are flat and tend to meet at 90° or 180°. In this case, grain growth slows down or even stops [3]. Lattice effects are decreased by changing to the triangular lattice [21], as in the present work. This is even more true at $T = 0$ [19]. However, it is possible to restore the free boundary motion in a square lattice by ''injecting a sufficient number of random steps onto the boundaries'', as reported by Holm and Battaile [16]. This can be accomplished, for example, by including second nearest interactions in Eq. (2) or by increasing temperature [16,23]. From this point of view, Liu et al. demonstrated that square and triangular lattices are equivalent when higher temperatures are used for the square lattice [1]. A comparison between MC simulations obtained with triangular and square lattice can be found in [24].

In order to compare the evolution of two different systems evolving at the same temperature, Fig. 5 reports the simulation results for two biphasic materials at $T = 0$. The first system is composed of 90% phase ''a'' (black squares) and 10% phase ''b'' (white squares), the second one is composed of 60% phase ''a'' and 40% phase ''b''. For both systems, the starting matrix (MCS = 0) and the matrix after 5000 MCS are shown. It is possible to see that, independently of the composition, the crystallization process induces a reorganization of the two phases, leading to the formation of phase ''b'' crystals within the phase ''a'' matrix, by means of atom migrations. In addition, the reorientation attempts within each phase, driven by the free energy reduction of the system, induces the formation of new domain structures. After 5000 MCS, the system has reached stability. For very long simulations (>25,000 MCS), in fact, the system shows negligible variations, as reported in Fig. 6.

The effect of increasing T is reported in Fig. 7. Since T is not related to real temperature, it is measured in arbitrary units. It

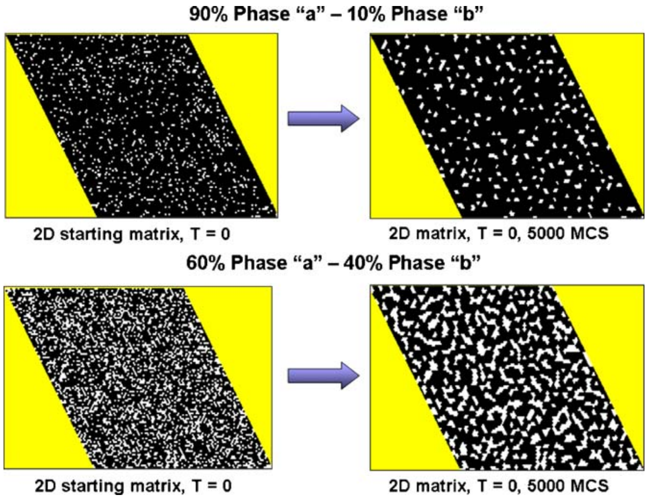


Fig. 5. Monte Carlo simulation: results for two different systems at $T = 0$. The corresponding starting matrix and the resulting one after 5000 Monte Carlo Steps (MCS) are reported. Being T non-related to real temperature, it is measured in arbitrary units.

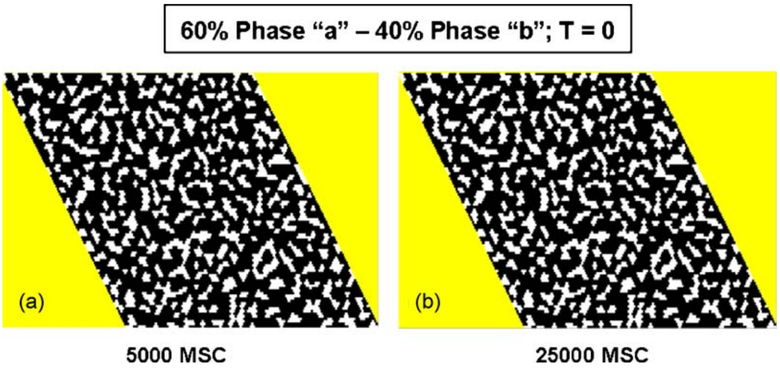


Fig. 6. Comparison between simulation results for 5000 and 25,000 MCS.

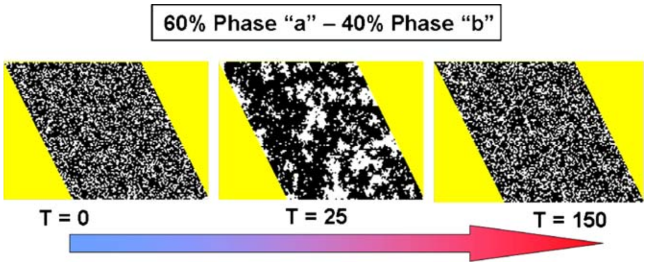


Fig. 7. The effect of increasing *T* for a 5000_MCS simulation.

should be noted that for a certain *T* range, the migration of phase ‘‘b’’ elements is favoured, which leads to the formation of large phase ‘‘b’’ domains within the phase ‘‘a’’ matrix. The ‘‘thermal’’ lattice vibrations provide the missing energy which is necessary to allow also those switching processes (see Eq. (3)) that locally increase the free energy, i.e. those switching processes associated with a $\Delta G > 0$. However, high energy transitions, i.e. with $\Delta G \gg 0$ are still prohibited. For large $k_B T$ values with respect to *V*, the lattice kinetics is such that all transitions are allowed, the system becomes chaotic and the formation of stable domains is not possible. Therefore, the system has no clots of phase ‘‘b’’ and it appears as an initial matrix, where the elements are arranged randomly.

4. Conclusions

An algorithm for the Monte Carlo simulation of crystallization and grain growth in biphasic-systems has been developed. The grain formation and growth is simulated by appropriate switching and reorientation attempts involving the matrix elements that describe the system. The crystallization in biphasic-systems with different volume fractions of the constituent phases has been simulated. In addition, the effect of temperature on the crystallization process has been discussed.

Although the proposed method describes the crystallization process from a macroscopic point of view, the algorithm contains several approximations, therefore a further development is under investigation. In particular, different Hamiltonians are under study with the aim to describe at best the grain growth kinetics and distribution. The main objective is to take into account the mobility of the considered elements, which varies, for example, whether the element is on a grain boundary or within a grain. Moreover, a specific experimental campaign would be desirable to evaluate accurately the energy parameters that describe the interaction between two sites belonging to the same phase or to two distinct ones.

References

[1] Y. Liu, T. Baudin, P. Penelle, Simulation of normal grain growth by cellular automata, Scripta Materialia 32 (1996) 1679–1683.
[2] M.P. Anderson, D.J. Srolovitz, G.S. Grest, P.S. Sahni, Computer simulation of grain growth—I. Kinetics, Acta Metallurgica 32 (1984) 783–791.
[3] D.J. Srolovitz, G.S. Grest, M.P. Anderson, Computer simulation of grain growth-V. Abnormal grain growth, Acta Metallurgica 33 (1985) 2233–2247.
[4] D.J. Srolovitz, G.S. Grest, M.P. Anderson, Computer simulation of recrystallization—I. Homogeneous nucleation and growth, Acta Metallurgica 34 (1986) 1833–1845.
[5] D.J. Srolovitz, G.S. Grest, M.P. Anderson, A.D. Rollett, Computer simulation of recrystallization—II. Heterogeneous nucleation and growth, Acta Metallurgica 36 (1988) 2115–2128.
[6] F.Y. Wu, The Potts model, Reviews of Modern Physics 54 (1982) 235–268.
[7] F. Mandl, Statistical Physics, 2nd edition, Wiley, 1991.
[8] M.P. Anderson, G.S. Grest, D.J. Srolovitz, Computer simulation of normal grain growth in three dimensions, Philosophical Magazine B 59 (1989) 293–329.
[9] J.E. Burke, D. Turnbull, Progress in Metal Physics, vol. 3, Pergamon Press, London, 1952, p. 220.
[10] M. Hillert, On the theory of normal grain growth, Acta Metallurgica 13 (1965) 227–238.
[11] A. Harun, E.A. Holm, M.P. Clode, M.A. Miodownik, On computer simulation methods to model Zener pinning, Acta Materialia 54 (2006) 3261–3273.
[12] A.D. Rollett, D.J. Srolovitz, R.D. Doherty, M.P. Anderson, Computer simulation of recrystallization in non-uniformly deformed metals, Acta Metallurgica 37 (1989) 627–639.
[13] A.D. Rollett, D.J. Srolovitz, M.P. Anderson, Simulation and theory of abnormal grain growth—anisotropic grain boundary energies and mobilities, Acta Materialia 37 (1989) 1227–1240.
[14] P. Zhu, R.W. Smith, Dynamic simulation of crystal growth by Monte Carlo method—I. Model description and kinetics, Acta Metallurgica et Materialia 40 (1992) 683–692.
[15] V. Tikare, J.D. Cawley, Numerical simulation of grain growth in liquid phase sintered materials—I. Model, Acta Materialia 46 (1998) 1333–1342.
[16] E.A. Holm, C.C. Battaile, The computer simulation of microstructural evolution, JOM 53 (2001) 20–23.

[17] E.A. Holm, G.N. Hassold, M.A. Miodownik, On misorientation distribution evolution during anisotropic grain growth, Acta Materialia 49 (2001) 2981–2991.

[18] R. Messina, M. Soucail, L. Kubin, Monte Carlo simulation of abnormal grain growth in two dimensions, Materials Science and Engineering A308 (2001) 258–267.

[19] P. Blikstein, A.P. Tschiptschin, Monte Carlo simulation of grain growth, Materials Research 2 (1999) 133–137.

[20] B. Radhakrishnan, T. Zacharia, Simulation of curvature-driven grain growth by using a modified Monte Carlo algorithm, Metallurgical and Materials Transaction A 26A (1995) 167–180.

[21] G.N. Hassold, D.J. Srolovitz, Computer simulation of grain growth with mobile particles, Scripta Metallurgica et Materialia 32 (1995) 1541–1547.

[22] Y. Kim, S.K. Hwang, M.H. Kim, S.I. Kwun, S.W. Chae, Three-dimensional Monte-Carlo simulation of grain growth using triangular lattice, Materials Science and Engineering A 408 (2005) 110–120.

[23] E.A. Holm, J.A. Glazier, D.J. Srolovitz, G.S. Grest, Effects of lattice anisotropy and temperature on domain growth in two-dimensional Potts model, Physical Review A 43 (1991) 2662–2668.

[24] Y.T. Liu, X.J. Guan, X.M. Shen, X.F. Ma, L.J. Wang, On the Monte Carlo simulation of normal grain growth, Acta Metallurgica Sinica (English Letters) 21 (2008) 282–288.