Computational Materials Science 92 (2014) 114–119

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

# A phenomenological approach to investigate nanocrystalline grain growth

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ARTICLE INFO

*Article history:*
Received 3 December 2013
Received in revised form 6 May 2014
Accepted 15 May 2014

*Keywords:*
Grain growth
Nanocrystalline microstructure
Grain boundary junctions
Junction drag effect
Phenomenological approach

ABSTRACT

A phenomenological approach to model the size effect observed during grain growth in nanocrystalline materials is presented. To that aim, the standard Monte Carlo Potts model is modified such that the mobility of grain boundaries depends on their triple junction distance. For initially very small grains growth kinetics are observed that are in agreement with experimental results, theoretical predictions and another simulation approach using a direct modification of the triple junction mobilities.



## 1. Introduction

The grain size of polycrystalline metals is the most important materials property since it influences nearly all mechanical properties like yield and tensile strength as well as toughness and ductility. An increase in grain size changes these properties, which has been making grain growth an important research topic for decades. However, unlike conventional micrometer-sized materials, metals and alloys of nanocrystalline size have quite different properties, like high values of hardness and superplastic behavior at low temperatures implying a size-effect. In particular, they are characterized by stable grain sizes even at relatively high temperatures and linear growth kinetics in contradiction to parabolic normal grain growth [1,2]. This is, of course, of great technological interest because an increase in grain size from nm to μm can result in a loss of essential materials properties making them unusable in applications.

One classical approach to investigate grain growth is the use of computer simulations representing a possibility to bridge the gap between experiments and analytic theories. This is especially valuable regarding the temporal development of microstructures, since they yield data not only about average grain size and associated size distribution but also about the morphology of grains and the topology of grain microstructures. Hence different mesoscopic simulation methods have been developed, which recreate grain growth numerically efficient and accurate and allow the observation of large grain ensembles over long time spans making statistical analyses possible. In particular, the Monte Carlo Potts model is based on the classical works of [3,4] and has been applied to, e.g., anisotropic and abnormal sub-grain growth, texture-controlled grain growth, and coarsening under the influence of second phase particles. However, recently it has also been shown in [5,6] that a modification of the standard Potts model by assigning each grain feature an own specific mobility enables the observation of the junction drag effect in nanocrystalline materials observing linear growth kinetics that change in later stages to parabolic behavior in agreement with experimental results [1,2], where it should be noted that particularly the paper from Paul and Krill [2] explained the anomalous growth kinetics (linear growth law and a broadening of the size distribution followed for long-time annealing by a narrowing of the distribution) by abnormal grain growth, but as we will see the same anomalous kinetics can also be obtained by triple junction controlled grain growth.

These considerations of a limited mobility of grain boundary junctions are based on the idea of the working group of Gottstein and Shvindlerman, who considered already in 1998 triple junction dragging and its influence on the von Neumann–Mullins-relation [7]. Their work was rooted in the idea that in nanocrystalline materials the influence of the triple points of a 2D grain network is non-negligible. In particular, the triple point mobility is considered to

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http://dx.doi.org/10.1016/j.commatsci.2014.05.026
0927-0256/

be different from that of the adjoining boundaries and influences not only the migration of the triple point but also the motion of the adjacent boundaries. To that aim, Gottstein and Shvindlerman derived a relationship between triple junction mobility and the forming dihedral angles (compare [8,9]). They concluded [10] that grain growth can be controlled by grain boundary junction mobility and the established structures are stable particularly for ultrafine grained and nanocrystalline materials. On the other hand, Barrales-Mora et al. [11] have shown that although the equations for the prediction of the growth rate of 2D grains under triple junction controlled grain growth are in good agreement with network model simulations, the initial microstructure has a very strong influence on the growth kinetics. In addition, an extension of the analytic growth laws as well as of the network model simulation to 3D [12] resulted also (in the average) in a good agreement. Deviations of the simulation results from the predicted kinetics were attributed to grains with geometries that were found to be extremely different compared to the generalized *N*-hedra—as originally defined by Glicksman [13]—that have been used to derive the analytic growth laws.

While a universal explanation regarding the stabilization of nanocrystalline grain structures and the number of factors influencing the grain boundary mobility has not been found yet, the size-dependence of the mobility of nano-sized grain boundaries is considered to be one of the most important effects. For further investigation, Streitenberger and Zöllner [14,15] considered grain growth as a dissipative process driven by the reduction of Gibbs free interface and junction energy. In particular, by attributing the observed size effect in nanocrystalline grain growth to a non-vanishing specific energy and a limited mobility of the grain boundary junctions (triple and quadruple junctions), an average growth law and a corresponding scaled grain size distribution function were derived for eight possible scaling states at small average grain sizes. Especially for triple line and quadruple point mobility limited grain growth, the corresponding scaled size distribution was found to be shifted strongly to small relative grain sizes describing a pseudo-steady state. The analytical size distributions of triple line and quadruple point limited grain growth compare very well with results from modified Monte Carlo Potts model [5,6]. In addition, in a very recent work Streitenberger and Zöllner [16] modeled triple junction controlled grain growth in 2D nanocrystalline polycrystals and thin films. For the case of triple junction drag controlled grain growth the influence of finite triple junction mobilities on metrical and topological properties was studied and analytical expressions of the self-similar grain size distribution were derived. The analytic results were found to be in very good agreement with modified Monte Carlo Potts model and front-tracking vertex dynamic simulations.

Although polycrystalline grain microstructures and therewith also grain growth is 3D in nature, nevertheless, two-dimensional grain growth is an important theoretical and computational concept that is used for analyzing basic properties and kinetics of grain coarsening as well as for describing grain growth processes in thin three-dimensional systems still today. For instance, in recent times, Barmak et al. [17] analyzed the growth kinetics of nanocrystalline Al and Cu thin films during annealing and found that triple junction drag can be considered as one of the possible causes of the observed grain growth stagnation. The analytical model of Streitenberger and Zöllner [16] describing 2D nanocrystalline polycrystals has been used also for a theoretical description of the experimental data obtained in annealed nanocrystalline thin films by Barmak et al. [17] yielding an excellent agreement.

In the following, a two-dimensional phenomenological approach to simulate the size effect observed during nanocrystalline grain growth is presented, where the mobility of grain boundaries depends particularly on their triple junction distance

## 2. A phenomenological implementation of the Potts model

While in principle the basic Monte Carlo Potts model is size-invariant, it has been shown in an earlier work by the author [5,6] that a modification of the Potts model assigning each grain feature its own specific mobility allows the observation of the size effect in nanocrystalline grain growth under triple and quadruple junction control.

In the standard Potts model [3,4] a grain microstructure characterized by a well-defined initial grain size distribution is mapped onto a quadratic 2D lattice with periodic boundary condition and eight nearest neighbors *nn*. Each lattice point (Monte Carlo Unit, MCU) comprises a certain but unassertive area of one grain. Following the standard definition the time unit of the simulation is one Monte Carlo Step containing *N* reorientation attempts, where *N* is the total number of lattice points. Each reorientation attempt works as follows: In the first step one lattice point is chosen at random, which is then given a new orientation chosen from all neighboring lattice points generating a new lattice state. For both states the energy is calculated using the Hamiltonian:

$$H = \frac{1}{2}\cdot\sum_{i=1}^{N}\sum_{j=1}^{nn}\gamma\cdot\left(1-\delta(Q_i,Q_j)\right), \tag{1}$$

where $\gamma$ is the specific grain boundary energy per unit length and measures the interaction of a MCU with all neighboring ones. It is a function of the misorientation angle between neighboring grains and can be calculated with the Read–Shockley-equation. The Kronecker delta function in Eq. (1) is equal to one if the neighboring orientations are the same and zero otherwise.

The final orientation of the selected lattice point is chosen with probability

$$p = \begin{cases} \frac{m}{m_{\max}}\frac{\gamma}{\gamma_{\max}}, & \Delta E \leqslant 0. \\ \frac{m}{m_{\max}}\frac{\gamma}{\gamma_{\max}}\cdot\exp\left(\frac{-\Delta E}{k_B T}\frac{\gamma_{\max}}{\gamma}\right), & \Delta E > 0. \end{cases} \tag{2}$$

$m$ is the grain boundary mobility given by the Huang–Humphreys-relation, $T$ is the simulation temperature and $k_B$ Boltzmann's constant. In particular, the constants $m_{\max}$ and $\gamma_{\max}$ describe the maximum values of mobility and energy, which take in a standard Potts simulation the value of one characterizing high angle grain boundaries. Hence, setting $m = m_{\max}$ and $\gamma = \gamma_{\max}$ yields a simulation of normal grain growth (compare, e.g., [18]).

In such a standard Potts model it is assumed that only the grain boundaries and their properties control the migration kinetics. The other structural elements of a grain network – triple and quadruple junctions – have no direct influence. Then again, as already mentioned a limited triple junction mobility is considered to be a main reason for the nanocrystalline growth kinetics. In particular, Gottstein and Shvindlerman [10] derived a principle expression giving the velocity of a boundary in terms of the intrinsic mobilities $m_{gb}$, $m_{tj}$, $m_{qp}$ of grain boundaries, triple and quadruple junctions

$$v = \frac{m_{gb}\gamma_{gb}K}{1+\frac{m_{gb}}{a m_{tj}}+\frac{m_{gb}}{a^2 m_{qp}}}, \tag{3a}$$

where $\gamma_{gb}$ is the specific boundary energy, $K$ is the curvature of the boundary, and $a$ is the boundary junction spacing. Introducing the intrinsic size parameters $\lambda = m_{gb}{:}m_{tj}$ and $\Lambda = m_{gb} : m_{qp}$, Eq. (3a) takes the simplified form $v = m_{eff}\gamma_{gb}K$ with an effective mobility

$$m_{eff} = \frac{m_{gb}}{1+\lambda/a+\Lambda/a^2} \tag{3b}$$