Scripta Materialia 109 (2015) 52–55

Contents lists available at ScienceDirect

Scripta Materialia

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

von Neumann–Mullins-type evolution equations for triple and quadruple junction controlled grain growth

Peter Streitenberger *, Dana Zöllner

Institute for Experimental Physics, Otto von Guericke University Magdeburg, Universitätsplatz 2, 39106 Magdeburg, Germany

ARTICLE INFO

Article history:
Received 20 May 2015
Revised 9 July 2015
Accepted 14 July 2015
Available online 18 July 2015

Keywords:
Grain growth
Nanocrystalline material
Junction control
von Neumann–Mullins-relation

ABSTRACT

The classical von Neumann–Mullins-law relates the area change rate of a grain in a two-dimensional polycrystalline microstructure to the number of neighboring grains yielding the well-known "*N*-6"-rule. In the present paper, we show that similar relations exist for the rate of size change as a function of number of neighboring grains for two- and three-dimensional polycrystalline grain microstructures under triple junction- and quadruple junction-control, which we find to be in very good agreement with simulation results.

Grain growth is generally based on the fact that polycrystalline materials are thermodynamically unstable. They undergo continuous coarsening driven by a reduction of the total Gibbs free energy of the system, where it is conventionally assumed that only the grain boundaries contribute to the reduction via the decrease of the total grain boundary area. In particular, for the simplest case of normal grain growth it follows that only two characteristic parameters enter the theory: namely the specific energy and mobility of the grain boundaries. As a result grain growth in polycrystalline materials always occurs due to the fact that individual grains either grow or shrink, and in case they do shrink they will surely vanish at some point reducing the total number of grains. This relaxation process takes place under geometrical and topological constraints of the grain network consisting of grain boundary faces, triple lines and quadruple points.

The fundamental relation between the rate of size change and the number of sides of two-dimensional soap froth derived by von Neumann in the 1950s [1] had been applied by Mullins to polycrystalline grain microstructures under normal grain growth in two dimensions [2] yielding what is known today as the classical von Neumann–Mullins-law,

$$\dot{A} = -m_{gb}\gamma_{gb}\left(2\pi - \frac{\pi}{3}N\right) = m_{gb}\gamma_{gb}\frac{\pi}{3}(N-6), \qquad (1)$$

where the area of a polygonal grain is given by A and the number of sides or neighbors of that grain and the number of triple points, respectively, is given by N. The parameter m_{gb} describes the grain boundary mobility and γ_{gb} the grain boundary surface tension. In particular, all grain boundaries are characterized by a unique value for the surface tension as well as by the same mobility in agreement with the uniform boundary model. It follows from Eq. (1) that all grains with more than six sides will grow and grains with less than six will shrink making six the critical number of neighboring grains N_c at which a grain will neither shrink nor grow. In particular, the von Neumann–Mullins-law holds for self-similar grain growth whenever the average grain area of the ensemble increases linearly with time, i.e., under normal grain growth.

Over the years, the von Neumann–Mullins-law has been verified for normal two-dimensional grain growth by experiments, theoretical considerations, and computer simulations (e.g., [3–5]). But also the effect of an additional driving force for grain growth on this relation has been considered for example in the works of Molodov et al. [6], where the drag effect of a magnetic field on microstructure evolution in non-ferromagnetic materials has been analyzed. However, especially in the last decade investigations of the influence of junction drag on grain growth kinetics have become increasingly important. Triple junction drag is the number one explanation for thermal stability of nanocrystalline materials (see overview [7]). While there is intense activity in this field of research regarding the production as well as the mechanical properties of nanocrystalline materials, statements regarding the grain microstructural changes during coarsening are generally limited. Nevertheless, two working groups have investigated this problem carefully.

* Corresponding author.
E-mail address: peter.streitenberger@ovgu.de (P. Streitenberger).

http://dx.doi.org/10.1016/j.scriptamat.2015.07.016
1359-6462/

Gottstein and Shvindlerman [8] analyzed the influence of triple junction dragging on the von Neumann–Mullins-relation based on the idea that in nanocrystalline and ultra-fine grained materials the influence of triple points of a 2D grain network is non-negligible considering that the triple point mobility is different from that of the adjoining boundaries (see also [9]). As a result they derived among others two separate functions for the rate of area change for grains with few ($N \leqslant 6$) and many ($N > 6$) sides. In addition to N, the area change rate depends also on the dihedral angle at the triple junction, which in turn depends strongly on the ratio of the mobilities of grain boundaries m_{gb} and triple junctions m_{tj} (compare also [10–12]). Also Barrales-Mora et al. [13] have shown that the effect of a limited mobility of triple lines on the volume rate of change can be implemented in 3D grain growth theories based on the average N-hedra approach introduced by Glicksman [14]. Based on the latter concept of average N-hedra Rios and Glicksman [15] considered in a very recent paper also the effect of grain boundary junctions in 3D.

The present authors [16] investigated for the case of pure triple junction drag the influence of finite triple junction mobilities on metrical and topological properties of a 2D grain network. We derived a self-similar expression of the grain size distribution as well as of the rate of size change showing that under pure triple junction drag the radius change rate depends uniquely only on N. The analytic results are found to compare very well with results from modified Monte Carlo Potts model simulations done by Zöllner and Rios [17].

In the present paper we show that the evolution equation of von Neumann–Mullins-type for 2D triple junction limited grain growth as it has been derived in [16],

$$\dot{R} = m_{tj}\gamma_{gb} 3\pi\alpha \frac{N - N_c}{N}, \tag{2}$$

is of more general validity than it is suggested by the linear Ansatz for the topological function $N = N(x)$ that has been used in [16]. The topological function $N(x)$ describes the relation between the number of neighboring grains N and the relative grain size $x = R/\langle R\rangle$, where R is the grain radius, $\dot{R}$ its rate of size change, and $\langle R\rangle$ the ensemble average. α is a numerical factor explained in [16]. In general, $N(x)$ can be approximated by a quadratic polynomial [18] $N(x) = c_2x^2 + c_1x + c_0$, where for two-dimensional triple-junction drag dominated grain growth as described by Eq. (2) $c_2 = 0$ [16].

In the following, similar forms of the evolution equation are presented for the cases of triple and quadruple junction limited grain growth in 3D polycrystals as they have been considered by the authors in [18].

An effective polygonal or polyhedral grain in a two- or three-dimensional polycrystal, respectively, can be described by an evolution equation according to Refs. [16,18]

$$R\dot{R} = m_{eff}\gamma_{gb}\left(\frac{R}{R_c} - 1\right), \tag{3}$$

where the driving force is the reduction of the total grain boundary energy γ_{gb}, and the critical grain size R_c is defined through $\dot{R}(R = R_c) = 0$. For a two-dimensional system $R\dot{R}$ is related to the grain area change rate $\dot{A} = 2\pi R\dot{R}$, and for a three-dimensional system to the affine grain volume change rate $V^{\dot{2}/3} = (2/3)(48\pi^2)^{1/3}R\dot{R}$. The effective mobility m_{eff} for a two-dimensional polycrystal is given by [16]

$$m_{eff} = \frac{m_{gb}}{1 + \frac{m_{gb}}{3\pi m_{tj}}\frac{N}{R}}, \tag{4}$$

where m_{gb} and m_{tj} are the finite mobilities of the grain boundaries and triple points, respectively. For a three-dimensional polycrystal the effective mobility is given by

$$m_{eff} = \frac{2m_{gb}}{1 + \frac{m_{gb}\sqrt{N}}{3m_{tj}R} + \frac{m_{gb}(N-2)}{4\pi m_{qj}R^2}}, \tag{5}$$

where m_{tj} and m_{qj} are the mobilities of triple lines and quadruple points of a grain in by a polyhedral network (cf., Eq. (4) in [18]). When the effective mobility m_{eff} in Eq. (3) is replaced by the grain boundary mobility m_{gb} and the critical grain size R_c is considered as grain size independent, $R_c = R_c(t)$, Eq. (3) corresponds to Hillert's approximation for normal grain growth [19]. However, even for normal grain growth R_c does depend also on the size of the considered grain, that is $R_c = R_c(t,R)$, reflecting a local spatial grain size correlation, which has been analyzed in [20,21] (see also [22–24]). As we will show in the following, this spatial grain size correlation can be expressed by the geometrical requirement for connectivity leading to a von Neumann–Mullins like representation of the various types of junction drag controlled grain growth.

Assuming that in a D-dimensional polycrystal a grain of size R is surrounded by N average N_c-hedra of equal size R_c each of which in turn has N_c neighbors. Connectivity of the polyhedral network (cf., e.g., [25,26]) requires that the considered grain of size R is connected with each of the surrounding average N_c-hedra of size R_c along a mutual shared grain boundary face, which for a given vertex-to-vertex distance a (cf., e.g., Fig. 1 in [18]) has a size in order of $A_D = \kappa a^{D-1}$, where κ is a dimensionless shape factor. Since the total surface areas S_D and S_D^c of the considered N-sided grain and the adjacent average N_c-sided grain, respectively, scale with their grain size as $S_D \propto R^{D-1}$ and, $S_D^c \propto R_c^{D-1}$, respectively, the area A_D of their mutual shared boundary face is given by $A_D \propto R^{D-1}/N = R_c^{D-1}/N_c$. Consequently,

$$\frac{R}{R_c} = \frac{N^{\frac{1}{D-1}}}{N_c^{\frac{1}{D-1}}}. \tag{6}$$

Inserting this relation into Eq. (3) yields the general expression

$$R\dot{R} = \frac{m_{eff}\gamma_{gb}}{N_c^{\frac{1}{D-1}}}\left(N^{\frac{1}{D-1}} - N_c^{\frac{1}{D-1}}\right), \tag{7}$$

describing the curvature term of the driving force purely by the number of faces N and a critical number of faces N_c. Although the reasoning, which leads to Eq. (6), is valid only for sufficiently large N, we show in the following that Eq. (7) represents a very good approximation not only for large but also for small values of N.

The quasi-stationary self-similar state of grain growth is characterized by separability of the evolution equation, that is $\dot{R} = \langle\dot{R}\rangle(t)\cdot G(x)$, where $G(x)$ is a dimensionless growth law that depends solely on the relative grain size $x = R/\langle R\rangle$ [19–23]. This implies also that N is only a function of the relative grain size, that is $N = N(x)$, and only one of the mobilities in Eqs. (4) and (5) can take a finite value while the remaining mobilities must tend to infinity [16,18].

In the case of two-dimensional grain growth, $D = 2$, Eq. (7) reduces for normal grain growth, $m_{gb} \ll m_{tj}$, to the well-known von Neumann–Mullins-expression:

$$R\dot{R} = \frac{m_{gb}\gamma_{gb}}{N_c}(N - N_c) \tag{8}$$

as shown to be valid by Monte Carlo Potts model simulations of normal grain growth for individual grains embedded in a constant surrounding (Fig. 1a, cf., also [17]) as well as for polycrystalline microstructures (Fig. 1b).

For triple junction limited grain growth, that is in case $m_{tj} \ll m_{gb}$, it follows:

$$\dot{R} = \frac{3\pi m_{tj}\gamma_{gb}}{N_c}\frac{N - N_c}{N} \tag{9}$$