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Zöllner, D.; Pantleon, W.

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Effect of boundary grooving on grain growth by Potts model simulations

D Zöllner and W Panteleon

Department of Civil and Mechanical Engineering, Technical University of Denmark DTU, Produktionstorvet 425, 2800 Kgs. Lyngby, Denmark

Abstract. Grain growth in thin films is a technologically important and scientifically fascinating topic: As the grain size influences many mechanical and optical properties of a material, any change in the grain structure and therewith in the grain size changes also the materials properties. There are many factors influencing coarsening of the microstructure such as grain boundary properties or the presence of particles; additionally the film thickness becomes rather important. It has been observed that when the grain size reaches the magnitude of the layer thickness, coarsening slows down and may even stagnate. For thin films, surface effects such as grain boundary grooving become important. In general, grooves are totally left out of implementations in the standard Potts model. In the current investigation, we introduce grain boundary grooves in the Potts model as physical entities. In the analysis, we show how the mere presence of such grooves reduces the driving force of grain boundary migration at the surface tremendously at any stage of coarsening.

1. Introduction

Coarsening phenomena such as grain growth have been a key field of study in materials science for more than 70 years. In the 1950s, Smith [1] established one of the first physically motivated grain growth theories. Other pioneers like Mullins [2] and Hillert [3] added to our understanding of grain growth by rationalizing growth rates and the occurrence of self-similar scaled grain size distributions, respectively. Additional knowledge accumulated on grain growth in the succeeding decades [4]. Starting in the 1980s, not only advanced experimental techniques but also newly developed computer simulation methods were developed that led to further insights.

In many grain growth studies, bulk materials are considered and surface and interfacial areas are neglected. For an analysis of thin films, however, surface effects — such as grooving or pinning of grain boundaries — are non-negligible. As the microstructure coarsens and the average grain size reaches the magnitude of the film thickness, grain growth slows down and may even reach stagnation. This phenomenon was observed in relatively thick [5] as well as very thin [6] metal films; Mullins [7] predicted that the formation of thermal grooves is the main reason for the stagnation of coarsening.

Grain boundary grooves form usually at elevated temperatures as atoms are able to move. While atoms may move in different ways such as diffusion in the lattice, diffusion at the surface or even evaporation from open surfaces, for grain boundary grooving atomic diffusion along the surface is most interesting. Driven by the surface and boundary energies, atoms move away from the intersection of the grain boundary with the open surface. A dent will be formed along the grain boundary and the removed material will deposit somewhere else on the surface. In this manner, local equilibrium is established between the grain boundary and the adjacent open surfaces.
This phenomenon as first described by Mullins [7] has been investigated thoroughly over the years. For example, Amram et al. [8] showed for thin nickel films that the shape of the grooves can be very different from that predicted by the classical Mullins model. Haremski et al. [9] studied grain boundary grooves in order to determine the relative grain boundary energies of nickel. Also grain boundary grooving in polycrystalline thin films was simulated using a phase-field model [10].

Analyses of grain microstructures in thin films is often done in two dimensions even today [11,12]. Such investigations assume by design that the grains in the microstructure are purely columnar. In reality, this will be the case quite rarely. In the recent past, three-dimensional computer simulations have shown how important it is to take the third dimension into account [13-15].

In the present investigation we analyze the effect of grain boundary grooves on grain growth in thin films by three-dimensional computer simulation. To that aim we implement for the first time physical grooves in a 3D Potts model and test their effect on grain coarsening at the surfaces as well as on the interior of the films.

2. Simulation method

2.1. Normal grain growth

For the current investigation, a lattice-based model, namely the Monte Carlo Potts model [16–18] is employed. A three-dimensional cubic primitive lattice is used to map a polycrystalline grain structure. Each lattice point or voxel represents a certain, otherwise unidentified region of a grain and is associated with the orientation of this grain. The lengths of each voxel along the coordinate axes is called Monte Carlo Unit (MCU). Ideally, there should be enough lattice points per grain to allow morphological characterization of each individual grain. Grain boundaries are defined between two neighbouring lattice points of unlike orientation. Triple junctions exist where three grains and, hence, three grain boundaries meet. In the Monte Carlo Potts model, individual voxels are tested randomly for potential orientation changes based on the configurational energy given by a Hamiltonian. A Monte Carlo Step (MCS) comprises the same amount of attempts as voxels in the lattice.

The underlying lattice must not affect the simulated growth kinetics. For this, the simulation temperature is of great importance, since it influences the grain boundary roughness as well as the angles between boundaries in triple junctions therewith ultimately affecting the growth kinetics as shown in [19,20]. While the simulation temperature is solely a technical parameter, (only) two real material parameters are relevant in the simulations, namely the grain boundary mobility $m$ and the grain boundary energy $\gamma$. The latter is the particular boundary energy per area and is employed by the Hamiltonian to determine the local energy of the microstructure on the lattice (which in turn determines the probability for orientation changes). Both, grain boundary energy and mobility, depend on the misorientation angle between the crystallographic orientations of the two adjacent grains. The grain boundary mobility influences the ultimate decision on the new orientation of a lattice point under consideration. Both, $m$ and $\gamma$ are only taken into account relative to their maximum values. In the current simulations, all grain boundaries are considered high angle boundaries having equal properties. Hence, energy and mobility are defined by their maximum values, $m/m_{\text{max}} = 1$ and $\gamma/\gamma_{\text{max}} = 1$, resulting in simulations of normal grain growth.

Commonly, periodic boundary conditions are utilized for such simulation to model the behaviour of bulk materials, assuming an endless continuation of the structure. This is assumed here as well in two of the three dimensions (left and right hand side in figure 1a). In contrast, the top and bottom layers of the underlying simulation lattice are regarded as free or open surfaces in the current study. Compared to all other lattice points within the bulk of a 3D cubic lattice which have 26 nearest neighbours, all points in the top and the bottom layer have only 17 nearest neighbour lattice points (5 first nearest neighbours, 8 second nearest neighbours, and 4 third nearest). The illustration in figure 1a depicts a two-dimensional section and lists the nearest lattice points in this section for three voxels as examples.

While such an implementation has been used in the past for simulations of grain growth in thin films under different conditions [13-15], none previous simulation ever contained grain boundary grooves. This raises the question, how to implement grooves as physical entities.
Figure 1. Sketch of a two-dimensional section through a digital 3D sample illustrating: a) the periodic boundary conditions at the vertical sample boundaries (left and right in the image) and free (top and bottom) surfaces at the horizontal sample boundaries. In addition, nearest neighbouring voxels are shown for three lattice points; b) the same section after introducing (small) grain boundary grooves. Different colours represent different orientations.

2.2. Introducing grain boundary grooves
As grain boundary grooves belong to the free space that surrounds the sample in an experiment, this feature must be transferred physically meaningful to the simulation. All voxels along grain boundaries at the top and bottom layer of the digital representation of a thin film are no longer associated with the respective grains, but rather with the free space around the digital sample as shown in figure 1b (coloured in white). Accordingly, the calculation of the local energy is changed in the simulation algorithm for a limited number of voxels. For lattice points that belong to a groove no attempt for a change is made as grooves are considered in the current implementation as fixed entities that do not change over time once introduced. If a lattice point neighbouring a groove is selected, the summation over the nearest neighbouring points changes (as shown in figure 1b) as grooves do not contribute to any change in local energy. This affects the probability for changing the orientation.

In the present investigation, it is assumed that grooves form spontaneously. They are introduced at a certain annealing time selected based on the progress of coarsening. The effect of two different types of grooves is investigated: small and large ones. The shapes of the grooves perpendicular to the surface and to the grain boundaries at the surface are shown in figure 2. Small grooves are realized by changing two lattice points adjacent to the grain boundary in the surface top or bottom layer, whereas large grooves result in a change of six lattice points distributed over two layers. After their introduction, the grooves are treated as fixed entities that neither change location, nor form anew.

Figure 2. Sketch of a section through a digital 3D sample perpendicular to the surface and a grain boundary showing from left to right a section containing the grain boundary before introducing a groove, a section that contains a small groove and a section that contains a large groove.

3. Simulation results
3.1. Normal grain growth
In order to analyse the effect of the uninhibited motion of grain boundaries at the surfaces without the presence of grooves, a thin film represented by a three-dimensional digital sample of size 400 MCU × 400 MCU × 50 MCU is investigated and simulations of coarsening under normal grain growth conditions are performed. Figure 3 confirms that the microstructure coarsens indeed and that boundaries intersecting a free surface align themselves perpendicular to them.
The following detailed analysis of grain coarsening is based on the average grain areas in individual layers parallel to the open surfaces. Analysing the average grain size at one of the surfaces (black curve in figure 4a) as well as in a layer in the centre of the film (green curve) reveals linear relations between grain size and annealing time after a short initial period of time. Consistent with earlier observations [13-15], the average grain size increases stronger at the surface than in the bulk. It must be noted that the sample presented here is still in its early coarsening stage. Even after the last simulation step (3000 MCS) only few grains (less than 20%) are columnar i.e. reach through the entire film. For very long annealing times it has been shown previously [13] that the coarsening will start to slow down approaching a new coarsening regime once approximately 80% of the grains are columnar.

Figure 4a illustrates how the average grains sizes differ in the different layers of the sample. While the average grain size is constant for the initial microstructure \((t = 0\text{ MCS})\), heterogeneity develops with increasing annealing time. The size profile adapts a bathtub shape. The average grain area increases faster at the surfaces than within the film while continuity of the grains is kept. Figure 4b also reveals statistical effects as the curves are not fully symmetric. Here it should be noted that for very long annealing times, this profile will flatten again, while the microstructures takes a columnar shape.
3.2. Grooving: Proof of concept
Introducing grain boundary grooves at an arbitrarily selected annealing time (here \( t = 500 \) MCS) results immediately in a different coarsening behaviour. An example of a coarsened microstructure at the top surface of the simulated thin film is given in figure 5. In figure 5a the microstructure is shown directly after introducing large grooves as described above. When the simulation is continued, it can be seen from figure 5b and c how only some grains grow beyond the fixed grooves into neighbouring grains. This is highlighted in the small magnified areas given below each image where even a new grain (in yellow) grows into the surface layer.

![Figure 5](image)

**Figure 5.** View on top layer of simulated sample including grooves that are marked in black: a) microstructure directly after generating the grooves; b) microstructure 100 MCS later; c) microstructure another 100 MCS later. For each image, the same small region is magnified below.

In the absence of grooves, undisturbed normal grain growth has been observed above with the average grain size following a linear relation with annealing time — even long after \( t = 500 \) MCS. These data are presented again in figure 6a as the black curve. The linear fit leads to slightly different values compared to figure 4a due to the limited time interval considered here.

![Figure 6](image)

**Figure 6.** a) Average grain area as function of annealing time at the surface layer in case of normal grain growth without grooves (black) and in the presence of grooves (blue) together with least-squares fits. b) Average grain area in individual layers parallel to the open surfaces as function of location, \( z \), within the film for six different annealing times either without (black) or with grooves (blue).
Introducing the grooves at $t = 500$ MCS yields immediately retardation of growth (blue curve in figure 6a), which persists within the observed time frame with the average grain area still increasing linearly in time. The fact that the initial grain size after adding the grooves becomes a bit larger than before is a result of a very small number of tiny grains that became deleted when introducing grooves. This minor effect does not influence the further coarsening.

The analysis above puts spotlight solely on the surfaces. Through the film thickness, a size profile with bathtub shape develops during coarsening as seen in figure 4b, where the average grain areas are larger close to the surfaces and increase — for early coarsening — somewhat faster than in the centre of the film. This can be seen in figure 6b again for normal grain growth between 500 and 700 MCS (black curves). What happens after introducing the grooves? The location dependent average grain areas after introducing the grooves are represented in figure 6b as blue curves. The growth in the interior of the film is basically unaffected and the coarsening progresses in the same manner as it does without the grooves at the open surfaces. In contrast, the average grain areas close to the surfaces are clearly affected showing retardation as expected from figure 6a. Simultaneously, the developed heterogeneity becomes slightly evened out.

For quantifying the retarding effect, average growth rates are calculated as rate of change in the average grain area in different layers depending on the location, $z$, within the film. For the simulation of normal grain growth the results mirror somehow the bathtub shape with larger growth rates closer to the surfaces as shown in figure 7a (black curve). For the interior of the film, coarsening yields the same results independent of the presence of grooves. In the presence of grooves (blue curve), the seven outermost layers, however, yield not only deviating behaviours, but also show growth rates clearly smaller than in the centre of the film confirming the strong retardation undoubtedly.

**Figure 7.** a) Average growth rate as rate of change in average grain area in individual layers parallel to the open surfaces for annealing times between 500 MCS and 700 MCS for coarsening without (black curve) and with grooves (blue curve). b) Growth rate of grain areas of individual grains at top surface as function of the number of their edges for coarsening without (top) and with grooves (bottom).

The growth rate has been in the centre of scientific attention for many decades—especially when it comes to the change rate of the area of individual grains. The von Neumann-Mullins-relation is one of the most fundamental laws of microstructural coarsening enabling a prediction of the kinetics of individual grains in polycrystalline metals and alloys [2,21,22]

$$\dot{A}(n) = m r \frac{\pi}{2} (n - 6)$$

where $\dot{A}$ is the two-dimensional area change rate of a grain and $n$ the respective number of edges. The derivation of this relation is based on the assumption of ideal coarsening in two dimensions.
Analysing the growth in area of individual grains at the surfaces of the thin films in the current investigation, which are not 2D entities, we still find for the thin film undergoing normal grain growth that the von Neumann-Mullins-relation is fulfilled (upper image in figure 7b). Even the critical number of edges distinguishing growing from shrinking grains is six. In contrast, once grain boundary grooves are introduced, the growth rate is no longer a function of the number of edges. Only few grains show temporal evolution at all. Of course, for longer annealing times, even these small changes will add up. In summary, the introduction of physical grooves exhibits a strong drag effect on the grain boundaries at the surfaces and affects also the regions within the thin films close to the surfaces.

3.3. Grooving: Simulation of long-time annealing
In order to investigate the effect of grooving to grain growth in thin films in a more general manner analysing also long-time annealing, we simulate a thin film of smaller thickness, namely 14 MCU. Due to this reduction, the size of the grooves is lowered to the small grooves as presented in figure 2.

As shown by the black curve in figure 8a, the average grain area increases linearly with annealing time after a very short initial period. The latter is a result of the initial microstructure not being fully relaxed yet. The observed linear dependence (up to 500 MCS) agrees with the results above and also with previous publications [13], where also a change to much slower coarsening for the late stage of annealing (here beyond 700 MCS) was observed. This change in growth kinetics is related to the formation of columnar grains. As seen from the black curve in figure 8b, the fast grain growth regime is coupled to a strong increase in the fraction of columnar grains, whereas the slow grain growth regime is associated with a more or less stagnant percentage of columnar grains close to 100 %.

![Figure 8. a) Average grain area at the top surface as function of annealing time for ideal grain growth (black curve) together with least-squares fits for early and late coarsening (dashed, red lines) as well as for grain growth in presence of boundary grooves (blue curves). For the latter, grooves are introduced after three different annealing times, namely at $t = 150$ MCS, 300 MCS, and 500 MCS. b) Associated fraction of columnar grains from same simulations.](image)

Once grain boundary grooves become introduced, one can notice from figure 8a that retardation sets in immediately. This is fully independent of the annealing time at which the grooves are introduced, here at $t = 150$ MCS, $t = 300$ MCS, or $t = 500$ MCS (blue curves). For the two simulations where grooving is introduced after 150 MCS or 300 MCS, coarsening progresses linearly just as in figure 6a, at least for a certain time span. For even longer annealing times, deviations from the linear relation occur which seem to be related to similar behaviour in the fraction of columnar grains. If grooving is introduced after 500 MCS (upper blue curve in figure 8a), the simulation starts at a point, where 90 % of the grains are columnar and the coarsening already started to slow down for normal grain growth. In this case, a fast transition to full stagnation is observed.
Here, we would like to emphasize the clear differences between long-time annealing behaviour for normal grain growth and grain growth controlled by grain boundary grooves: For normal grain growth the late stage of coarsening is associated with a slow coarsening of the microstructure; the number of grains decreases, while the fraction of columnar grains stays close to 100%. In contrast, grain growth controlled by grain boundary grooves results in a full stagnation of coarsening, which is reached in the current study for the case, where grooves are introduced at the stage with 90% columnar grains.

4. Summary and conclusion
In the present investigation, grain boundary grooves have been introduced for the first time in the 3D Monte Carlo Potts model for grain growth. The simulation algorithm is adopted such that at the top and bottom layer of the digital thin film voxels along grain boundaries are associated with the free space that surrounds the digital sample. The mere presence of such grooves reduces the driving force of boundary migration at the surfaces tremendously at any stage of coarsening. The effect sets in instantly. The interior of the films coarsens unaffected by the grooves, whereas coarsening rates at the surfaces are not only smaller for the grooved surfaces compared to the surfaces without grooves, but even smaller than the coarsening rate in the interior of the films. Grooves can even result in full stagnation of coarsening highlighting the importance of taking into account the effect of grain boundary grooving on grain growth.

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