Serial sectioning of grain microstructures under junction control: An old problem in a new guise

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Abstract. In the present work the importance of 3D and 4D microstructure analyses are shown. To that aim, we study polycrystalline grain microstructures obtained by grain growth under grain boundary, triple line and quadruple point control. The microstructures themselves are obtained by mesoscopic computer simulations, which enjoy a far greater control over the kinetic and thermodynamic parameters affecting grain growth than can be realized experimentally. In extensive simulation studies we find by 3D respectively 4D microstructure analyses that metrical and topological properties of the microstructures depend strongly on the microstructural feature controlling the growth kinetics. However, the differences between the growth kinetics vanish when we look at classical 2D sections of the 3D ensembles making a differentiation of the controlling grain feature near impossible.

1. Introduction

Today there still exist many gaps in our fundamental understanding of polycrystalline microstructures and in predicting their behavioral changes due to recrystallisation and grain growth. Closing this gap is of utmost importance since the grain size of a material determines its properties. Common stereological 2D investigations [1], e.g., using the mean linear intercept method, yield information on the average grain size, but under certain assumptions regarding the morphology of grains. Hence, only the 3D data provide the necessary metrical and topological properties of polycrystals. To that aim, in recent years much effort has been put into the global task to develop new, advanced materials, which has led to new characterization techniques enabling materials scientists and engineers to analyze bulk materials and thin films in 3D and 4D by X-rays, electrons, and neutrons with high resolutions in real time [2].

An analysis of grain microstructures by means of focused ion beam (FIB) tomography allows the creation of serial sections in an automated manner. After reconstruction the shapes and sizes of individual grains in the network can be described (compare, e.g., [3]). The combination of a focused ion beam and scanning electron microscope with the aim of a tomographic orientation analysis is already developed for about 10 years, but the use in the nanocrystalline grain size range, where the grain boundary junction can control the microstructural evolution, is not trivial as it leads the 3D tomographic Electron Backscattering Diffraction method (3D EBSD) method in conjunction with FIB sectioning to their resolution limit concerning the determination of the local orientation [4]. Then again, while recently the spatial resolution in 3D for the tomographic EBSD was about 50×50×50 nm^3 [5], today resolutions well below 50 nm (see [3]) can be achieved using low-voltage methods. For example Balach et al. [6] succeeded in generating 10 nm thin slices in FIB–SEM tomography experiments.

Experiments in nanocrystalline iron [7] have shown that microstructures with an average grain radius of well below 100 nm show anomalous behavior (e.g., a linear average growth law). Hence, such microstructures are important to investigate more closely. Using

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mesoscopic computer simulations [8] and analytic mean-field theories [9] it has been found that such behavior can be associated with triple junction drag leading among other things to a very strong increase in small grains. With an average grain size of less than 100 nm as in the experiments this yields a high number of small grains with radii as small as 10 nm. These can, however, not be portrayed with the resolution necessary to describe their morphology correctly. Hence, for such nanocrystalline grain microstructures we still might have to use stereological 2D investigations.

In the present work we study polycrystalline grain microstructures obtained by grain growth under grain boundary, triple line and quadruple point control. In extensive simulation studies we find by 3D respectively 4D microstructure analyses that metrical and topological properties of the microstructures depend strongly on the microstructural feature controlling the growth kinetics. Those differences between the growth kinetics become blurred when we look at classical 2D sections of the 3D ensembles making a differentiation of the controlling grain feature very difficult.

2. Grain growth under grain boundary junction control

Unlike conventional materials, metals and alloys of nanocrystalline size have quite different mechanical properties like high values of hardness, yield and fracture strength and superplastic behaviour at low temperatures implying a size-effect. They show stable grain sizes even up to relatively high temperatures and linear or even exponential growth kinetics in clear contradiction to parabolic normal grain growth [7, 10]. Such investigations of the stability of nanocrystalline materials during grain growth are, of course, of intense technological interest because an increase in grain size from nm to μm can result in a loss of important materials properties making them unusable in applications. Already in 1997 Malow and Koch [11] summarised significant works concerning the stabilization of nanocrystalline grain structures in many materials and the number of factors influencing the grain boundary mobility in nanocrystalline alloys, like grain boundary segregation, solute drag, pore drag, second phase (Zener) drag and chemical ordering. A universal explanation has not been found yet, but the discussion is still on.

In particular, Gottstein and Shvindlerman [12-14] proposed that grain growth can be controlled by grain boundary junction mobility. The established structures are rather stable, in particular, for ultra-fine grained and nanocrystalline materials. Streitenberger and Zöllner [9, 15] considered grain growth as a dissipative process driven by the reduction of Gibbs free interface and junction energy. A general grain evolution equation has been derived separating into nine types of growth kinetics. The corresponding self-similar grain size distributions were in agreement with first results from modified MC simulations considering size effects in triple and quadruple junction limited grain growth. Details on the simulation method as used in the present work can be found in [8, 16].

3. Evolution of the average grain size: 3D versus 2D measurements

If the grain microstructural evolution is controlled solely by the mobility of one particular structural feature (grain boundary, triple line or quadruple point) it has been shown [8, 9, 14] that the average growth law of the 3D structure follows in each case a unique time law. Under grain boundary (gb) control the classical parabolic growth law, $\langle R\rangle \propto \sqrt{t}$, holds (compare Fig. 1a). Under triple line (tl) control (Fig. 1b) a linear growth law results, while grain growth controlled by the quadruple points (qp) yields the exponential increase in annealing time as shown in Fig. 1c. In each case it can be seen that there is also an initial period of time that is not described by the respective laws depending on the initial microstructure. In addition, for each of the three cases the temporal development of the average grain size calculated from 2D sections, $\langle r\rangle(t)$, is also shown, from which it is evident that the growth laws follow the same general behavior as their 3D counterpart, while showing fluctuations that are due to the limited number of observed grains in the sections. Hence it seems that the microstructural feature controlling grain growth can be deducted from the observation of the growth law in its 3D or 2D version.

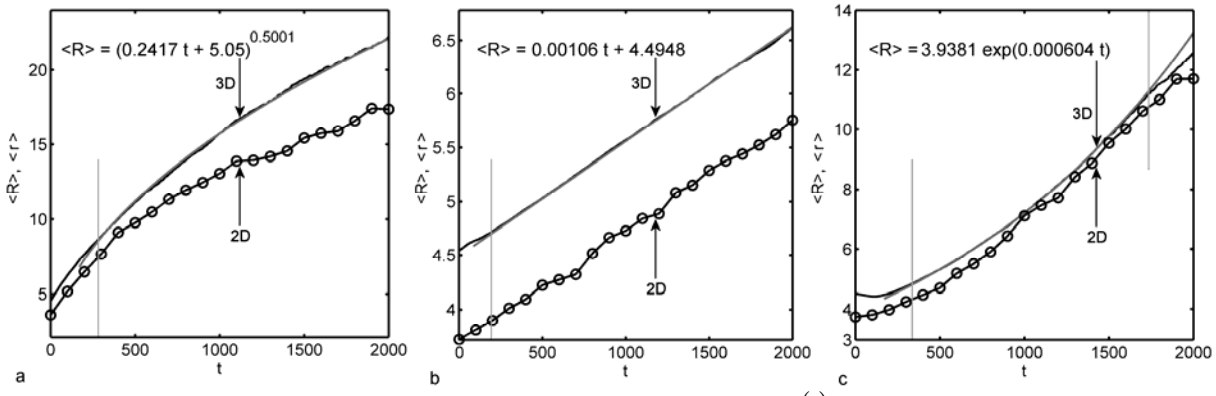

Figure 1. Temporal development of average 3D grain size $\langle R\rangle(t)$ and grain size in 2D sections $\langle r\rangle(t)$ for grain growth under: a – grain boundary control; b – triple line control; c – quadruple point control.

However, this is not entirely true as Figure 2 shows, where the three kinetics (gb control – circles; tl control – squares; qp control – triangles) are shown again as $\langle r\rangle(t)$, only now each together with a linear least-squares fit. It is evident that due to the limited number of measured data points a linear relation gives a good representation independent of the microstructural feature controlling the growth kinetics allowing no conclusions!

More frequent or longer observations can solve this problem, and although this is particularly important, it is not always possible to detect more observation times, since, e.g., for long time annealing the growth kinetics changes for example under triple line control from linear to parabolic (see [8]) restricting the observation range.

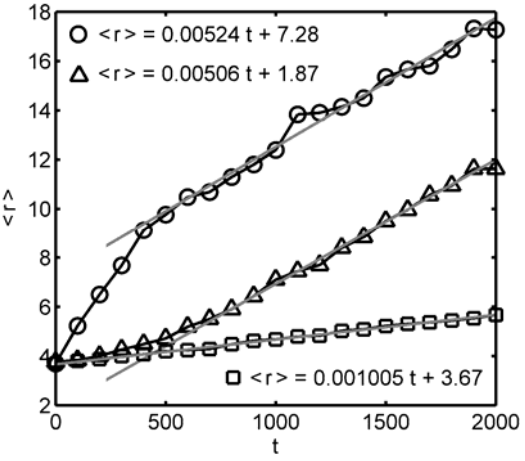

Figure 2. Development of average grain size in 2D sections for grain growth under: gb control (circles), tl control (squares), and qp control (triangles).

In order to allow nevertheless a reliable deduction of the microstructural feature that is in control of the growth kinetics, we show in the following that both, the metrical and topological properties of the grain network have to be investigated very thoroughly.

4. Grain size distribution in sections and real

Each of the above growth kinetics of a 3D polycrystal (as presented in Fig. 1) is associated with a distinct scaled grain size distribution $f(x)$ [8, 9], where x is defined as grain radius of a grain divided by the average radius of the ensemble. This is shown indeed in Figure 3a-c, where it can be seen that grain growth under limited triple line mobility with a linear increase in the average grain size (Fig. 3b) as well as under a limited quadruple point mobility with exponential growth law (Fig. 3c) are characterized by a remarkably high number of small grains compared to normal, parabolic grain growth (Fig. 3a). All three types of grain size distributions are found to be self-similar (i.e. time-independent) and in agreement with theoretical predictions (black lines in Fig. 3a-c; compare [9, 15]).

Then again, it is well known that sectioning of 3D polycrystals results in deviating grain size distributions compared to their 3D counterpart. For normal grain growth the scaled size distribution $f(y)$, where y is the scaled grain size of the section, is shifted to smaller grain sizes, while the distribution becomes broader and less peaked (Fig. 3d). Similar changes can also be observed for triple line controlled grain growth (Fig. 3e) and grain growth under quadruple point control (Fig. 3f). However, only the latter is distinctively different from normal grain growth allowing clear conclusions regarding the associated exponential growth

regime. The differences between the distributions in Figs. 3d and 3e are basically negligible giving us nearly no indication, which distribution belongs to parabolic normal grain growth and which to linear triple junction controlled growth. The only real difference is the maximum grain size: Under grain boundary control we can observe grains as big as 2.5 times the average size, whereas under triple junction control grains growth as large as 3.0 times the average size giving the distribution (Fig. 3e) a slightly larger tail.

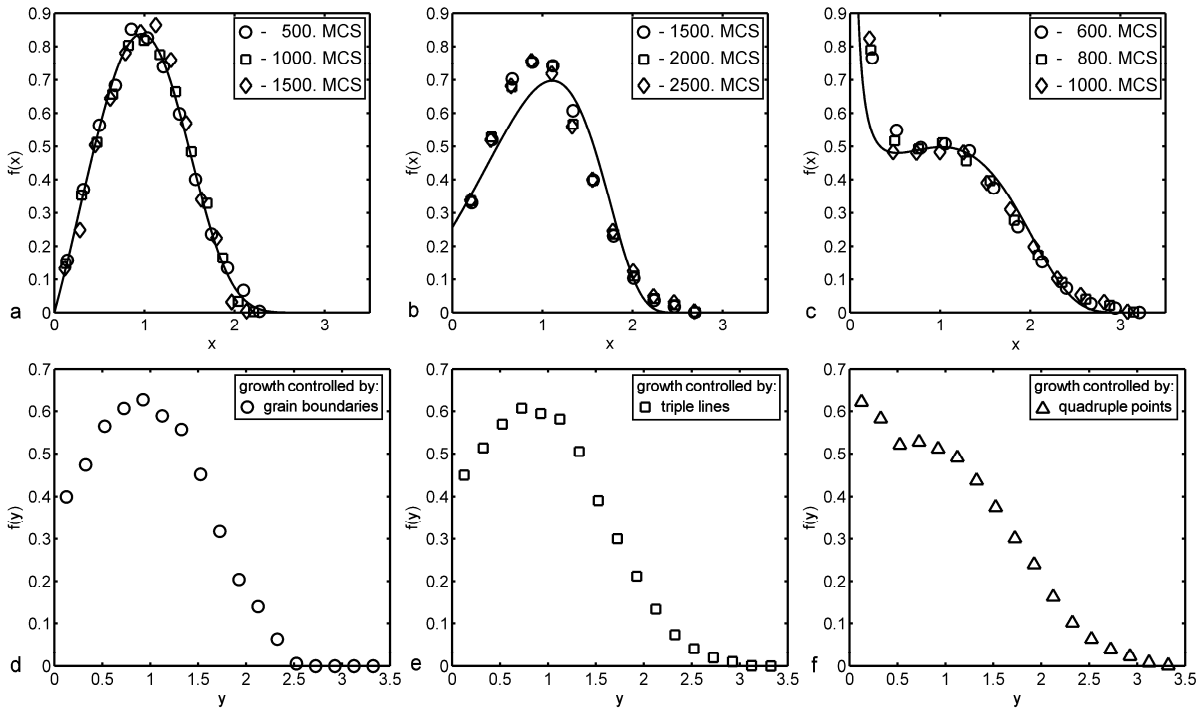

Figure 3. Relative grain size distributions: a – 3D normal grain growth; b – 3D triple line controlled growth; c – 3D quadruple point controlled growth; d – 2D sections from normal grain growth; b – 2D sections from triple line controlled growth; c – 2D sections from quadruple point controlled growth.

5. Topological investigations of 2D sections

In addition to metrical properties, polycrystalline grain boundary networks are also characterized by topological properties like the number of faces and edges of the grains. It is shown particularly in Figure 4a that the grain size *y* of the grains in the 2D sections is strongly correlated to the number of edges.

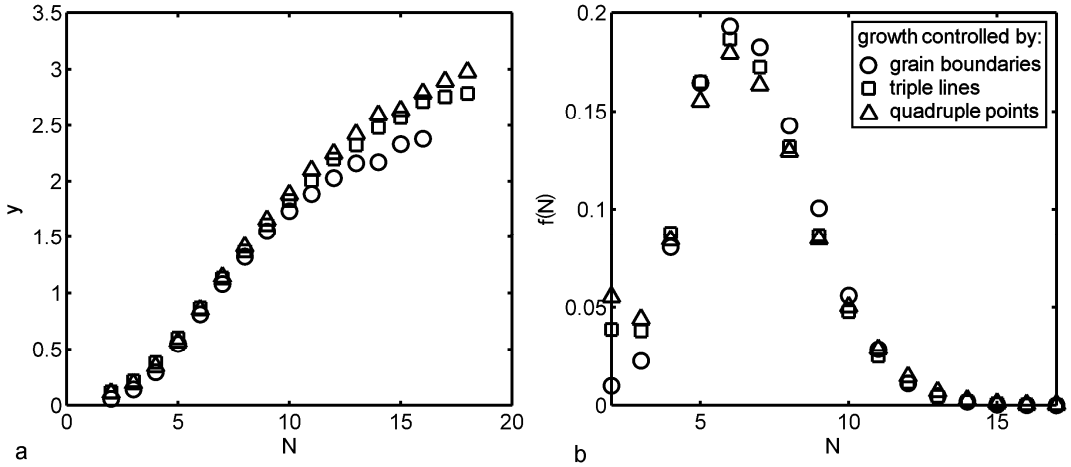

Figure 4. Topological properties of the grain networks: a – Relative grain size versus number of edges (averaged *y*-values for each edge class); b – Distribution of number of edges.

While it has been shown previously in [8] that the number of faces as a function of the grain size of the 3D microstructures allows a clear deduction of the microstructural feature that is in control of the growth kinetics, Fig. 4a shows that the 2D sections show virtually no differences for small and average grains. Only for large grains with many faces the triple and quadruple junction controlled kinetics differ significantly from normal grain growth. Then again there exist only very few of such large grains as the distributions of the number of edges show in Fig. 4b. In contrast the latter also illustrate the strong increase in the number of grains with few faces.

All in all, it can be concluded that while the metrical and topological properties of the 3D polycrystalline grain structures depend strongly on the microstructural feature controlling the growth kinetics (see, e.g., [8]), it is shown in the present work that those differences between the three growth kinetics become blurred when we look at classical 2D sections of the 3D ensembles. In particular, we have shown that due to a limited number of measured annealing times the average growth law can be described by a linear relation independent of the microstructural feature controlling the growth kinetics, which is in strong contrast to the 3D growth laws. Only from a combined observation of metrical and topological properties of the grain networks a reliable deduction of the microstructural feature that is in control of the growth kinetics can be drawn.

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