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GRAIN BOUNDARY, TRIPLE JUNCTION, AND QUADRUPLE POINT GRAIN GROWTH DYNAMICS

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Abstract

Grain boundaries, triple junctions, and quadruple points are the geometrical elements in 3-D polycrystalline networks, where these elements have co-dimensions 1, 2, and 3, respectively. Grain boundary mobility usually controls network kinetics in materials with "large" grain sizes. Nonetheless, there are studies concerning the effects of these network elements on the self-similar distributions of scaled grain size and number of faces per grain. This study applies a polyhedral representation of grains that the authors developed previously to obtain LSW-like (self-similar) behavior for distributions with three rate-limiting situations, viz., where the kinetics of network evolution is controlled by boundary, triple junction, or quadruple point mobility. The self-similar network states so obtained yield important implications concerning details of polycrystalline evolution, such as grain growth "trajectories", grain lifetimes, and their volume-time behavior.

Introduction

Polycrystalline grain growth, which is topological similar to soap bubble coarsening, was believed for a long time to be controlled solely by grain boundary (GB) mobility and curvature. Later, the importance of triple junctions (TJ) and quadruple points (QP) was recognized [1]. TJs and QPs become especially important as the grain size decreases to the nanoscale. Figure 1 illustrates how, at large (conventional) length scales, the density of GBs dominates grain networks. This situation changes as the network length scale decreases and a certain threshold TJ density is reached. At extremely small length scales, the density of QPs would eventually dominate.

Figure 1, however, depicts a static picture of the situation. When one takes into account grain growth *dynamics*, then the mobilities of GBs (m_{GB}), of TJs (m_{TJ}), and of QPs (m_{QP}), must be taken into account. Gottstein and Shvindlerman reconciled these additional kinetic factors and proposed a kinetic relationship between the grain boundary velocity, G, and its driving force, P, weighted by the scaled mobilities o all the network elements,

$$G = [1/m_{GB} + 1/(\lambda m_{TJ}) + 1/(\lambda^2 m_{QP})]^{-1} P \qquad (1)$$

Equation (1) shows that both mobility and length scale, λ, play a role in determining the boundary speed. For example, if the three network mobilities are equal then the length scale would provide the controlling kinetic factor, as shown in figure 1. By contrast, if one of the scaled mobilities, say λm_{GB}, is much smaller than the other two, as occurs in conventional

grain growth, then that slow step controls the network kinetics, and equation (1) reduces to $G=\lambda m_{GB}P$.

Rios and Glicksman [2] showed in a recent paper that long-term self-similar distributions of grain size and number of faces develop that correspond to situations controlled by one of the scaled mobilities, m_{GB}, λm_{TJ}, or $\lambda^2 m_{QP}$ that became the controlling kinetic factor. In what follows these three kinetically limiting cases will be referenced as GB-, TJ- or QP-controlled grain growth.

In this paper we first revisit the main results from Rios and Glicksman, and then extend their discussion drawing wider interpretations of their results.

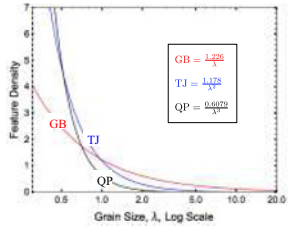

Figure 1: GB, TJ, or QP density as functions of the network length scale, chosen as the vertex-to-vertex distance, λ.

Self-similar distributions

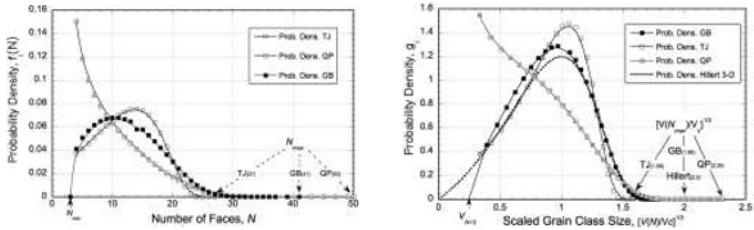

Figure 2: **left**, number of faces distributions (NFDs), and **right**, grain size distributions (GSDs) for the three network kinetic limits controlling polycrystalline grain growth: GB, TP, QP. Curves are the continuous self-similar distribution functions, where symbols designate the *discrete* probability densities for integer face classes.

Figure 2 shows the self-similar distribution of number of faces (**left**), and the distribution of grain sizes (**right**) obtained earlier by Rios and Glicksman using regularized polyhedral grains. Hillert's distribution for spherical grains is included for comparison in Fig. 2 (**right**), and nearly coincides with the polyhedral solution for GB-mediated kinetic control, as is expected from previous work [3]. Hillert's distribution does not normally show agreement with computer simulations [4]. Similarly, the number of faces distribution derived by employing Hillert's assumption of a constant vertex-to-vertex distance does not provide agreement with computer simulations either [3]. Consequently, close agreement between TJ- and QP-distributions derived here from computer simulations is also not expected. This was confirmed when the results shown above are compared with recent Monte Carlo simulations conducted by Zöllner [5].

There is, however, an important aspect for which the present analytical solutions agree well with computer simulations. This concerns the fact that GB-, TJ-, and QP-limited network kinetics *do possess* long-term, analytical, self-similar solutions independent of the controlling kinetic mechanism. We note that each controlling mechanism produces its own kinetic growth law and each is distinct from the others. Nonetheless, all three kinetic cases exhibit analytical self-similarity and this affine behavior is also found in computer simulations. From these results one may draw a tentative conclusion: *self-similarity (the affine state) is an intrinsic property of evolving polycrystalline networks.* Moreover, self-similarity can be attained independent of the controlling kinetic mechanism. Of course, the form of each affine distribution is determined by the corresponding controlling kinetic mechanism and each will be unique.

Grain trajectories

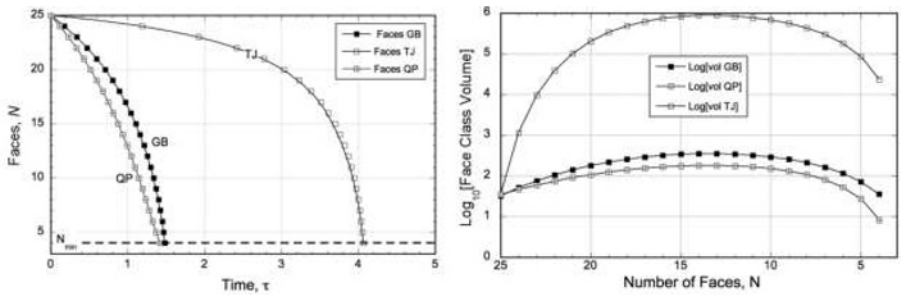

Figure 3: (**left**) mean faces loss trajectories as a function of time; (**right**) volume trajectories (log-scale) through face-number *N*-space, for *average* network grains initially having 25 faces, and evolving under each case of kinetic network control.

Figure 3 presents the expected grain faces loss trajectories, that is, how topological (number of faces) and how metrical (mean grain volumes, *V*) evolve. Figure 3 (**left**) shows how the number of faces of the grains evolve as they are lost by topological reactions. In all cases the number of faces per grain always decreases with time. Actually, it is possible to show that the number of edges per grain and number of vertices per grain also decrease, following, as they must, the decrease in number of faces per grain. Figure 3 (**right**) shows the expected volume evolution versus the number of faces. The volume for each face class always increases for grains with number of faces $N \geq 14$ but decreases for number of faces $N \leq 13$. All three volume curves pass through a broad maximum at the same critical *N*-value, $N_c \sim 13.4$. The reason for this is that between $N=14$ and $N=13$ the grain faces reverse the sign of their curvature. For $N \geq 14$ their faces are concave, and the grains increase their volume. In contrast, for $N \leq 13$ grain faces are convex, and, therefore, grains lose volume. Glicksman and Rios [6] demonstrated that a regularized polyhedral grain with $N=13.397\ldots$ faces has flat faces, and is the 3-D homolog of a six-sided grain in 2-D that neither increases or decreases its area. A 3-D grain with $N=13.397\ldots$ faces lacks any driving force either for growth or shrinkage. Moreover, this kinetic feature of the polyhedral face number does not depend on whether the rate controlling *kinetic* feature is the GB, TJ, or the QP mobility. Any evolving polycrystalline network *must* always comply with this general topological result: change of sign of the *driving force* for grain growth/shrink at $N_c \sim 13.397\ldots$ faces. The main result brought out in Figure 3 (**right**) is that when the self-similar state is finally achieved, *grains,*

on average, lose faces—even as they grow. This apparent paradox is resolved provided one considers the possibility of two independent processes resulting in grain volume increases or decreases, namely, 1) metrical and 2) topological changes.

This idea is best understood by writing the total differential of a grain's volume change, dV, as

$$dV(N,\lambda)=\left(\frac{\partial V}{\partial N}\right)_{\lambda} dN+\left(\frac{\partial V}{\partial \lambda}\right)_{N} d\lambda \qquad (2)$$

The first term in equation (2) represents the differential volume change when a grain loses or gains faces. The volume decreases when a grain loses a face during self-similar evolution. Therefore, the first (topological) term in equation (2) remains negative as face losses occur. Conversely, the mean vertex-to-vertex distance tends to increase over time, so that the second (metrical) term in equation (2) remains positive. Whether a particular polyhedral grain's volume increases or decreases volume with time depends on the interplay of the two independent terms in equation (2). This tendency will be discussed in the next section.

A related question that arises is precisely how self-similarity is maintained if grains are on average losing faces. The answer lies in the fact that self-similar distributions deal statistically with grain *classes* not with individual grains. Consider the class of grains with N faces and volume $V(N, \lambda)$. When a grain from the class with N+1 faces loses a face it joins the lower face class of grains with N faces. Since this transition is also accompanied by an increase in the vertex-to-vertex distance over time the volume of this "new", or added, grain with N faces will on average develop a larger volume, $V(N, \lambda+\Delta\lambda)$. In this manner the volumes of grains in *all* face classes increase, and eventually self-similar evolution results. Obviously, if the volume of each class increases over time, some grains must disappear, as the total network volume must be conserved. We discuss this important constraint in the next section.

It is also worth mentioning that a recent study on the transient states developed between an initial distribution of Voronoi polyhedra and the final self-similar distribution shows that if one is *not yet* in the self-similar state, then it is possible for a grain to gain faces [7]. Again we stress that the behavior of grains during self-similar growth is identical and independent of the controlling kinetic mechanism. Again, one may draw the conclusion that this consistent network behavior is a characteristic resulting from the long-term evolution of polycrystalline networks and does not depend on the network element controlling growth.

Metric and topological contributions

Figure 4 captures the detailed behaviors of grain volume rate of change with number of faces for the three different kinetic limits discussed herein. The total rate of volume change is decomposed into their topological and metric contributions as indicated in equation (2). As expected, the rate of change in the three limiting network kinetic cases considered is always equal to zero for Nc=13.397… Nonetheless, even grains with a number of faces very close to N=13.397…, N=13 or N=14, may suffer considerable topological and metric changes of opposite signs. These countervailing changes combine to produce small rates of change of the total volume.

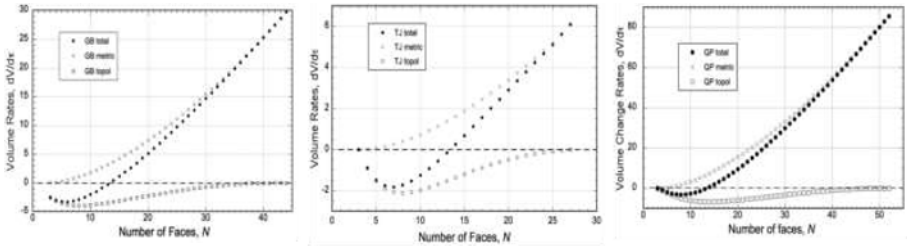

Figure 4: Metric and topological contributions to volume change rates: (**left**) GB-control; (**center**) TJ-control; (**right**) QP-control.

The most evident feature of the three graphs in figure 4 is that the metrical component is always positive, and the topological component is always negative. Fundamentally, this uniform behavior stems from the mathematical structure of the LSW methodology. In one of the intermediate steps of generating the self-similar solution we obtained an expression $dN/d\ln\lambda = U(N)$. The function $U(N)$ depends on two network parameters, one of which, is the constant α, related to the temporal growth law. For example, for GB control one obtains the well-known parabolic law, where $d\lambda^2/dt$ is equal to a rate constant determined by α. The second parameter, N_{max}, is the maximum number of polyhedral grain faces allowed in the self-similar state. In order to determine these two network parameters, the LSW method requires that the function $U(N)$ possess a *double* root at N_{max}, namely, $U(N_{max})=0$ and $dU(N_{max})dN=0$. The mathematical solution subject to these constraints yields a positive value for α, which combined with N_{max} insures that $U(N)<0$ for $N<N_{max}$. One can rewrite the expression above considering that $U(N)<0$: $(dN/dt)/(d\ln\lambda/dt)<0$. Owing to the fact that $\alpha>0$ all three kinetic mechanisms require that $d\lambda/dt>0$ so that $dN/dt<0$. In brief, the parabolic constant being positive insures that λ always increases, and $U(N)<0$, insures that N always decreases. These mathematical features of the LSW analysis for self-similar grain network evolution show why the topological component is always negative, i.e., $dN/dt<0$, whereas the metrical contribution is always positive, i.e., $d\lambda/dt>0$.

Another conclusion is that the number of grains per unit of volume must decrease during 3-D network grain growth, independent of the rate-controlling kinetic mechanism. Therefore, the mean grain volume must increase in every face class of grains. This feature is accomplished through the topological disappearance of grains. A related topological fact, not often mentioned, is that when a grain finally disappears, its *faces* also disappear. As grains leave the distribution, the faces of those grains are redistributed throughout the network via topological reactions with neighbor grains. The volume of the lost grains is also redistributed among the network's remaining grains, so that all grain *classes* exhibit yet larger average volume, even among face classes for which member grains shrink on average. For example, consider the class of grains with only 10 faces. As time passes fewer grains populate this class, which exhibits a larger average volume, even if some individual member *grains* within this class lose faces and decrease their volume. Similarly, the interfaces between grains that disappear are shared in the remaining network grains that increase their individual interfacial areas even as the total interfacial area and energy of the network decreases.

Finally, we emphasize that this discussion entails *mean* metric and topological values. Computer simulations, by contrast with this analysis, also expose *stochastic* aspects of grain growth associated with network evolution, which, in fact, exhibits a broad range of volumes.

Conclusions

The evolution of a polycrystalline network, conceptualized as a collection of space filling trivalent polyhedra was studied by analytical methods for three different kinetic controlling mechanisms corresponding to the scaled mobilities of the geometric elements of the network: grain boundaries, triple junctions and quadruple points. That analysis outlined above permits one to draw the following conclusions:

- The self-similar state is an intrinsic property of evolving quadravalent polycrystalline networks.
- The polycrystalline network reached a self-similar state independently of the controlling *kinetic* mechanism, namely, the GB-, TJ- or QP-mobilities.
- Nonetheless, the self-similar distributions of sizes or face numbers depend on the rate controlling kinetic mechanism.
- The grain trajectories were also similar for the three controlling mechanisms, but, of course, the numerical values depend on the controlling mechanism.
- All three curves of grain volume against number of faces, figure 3 (**right**), pass through a maximum at the critical face number value N_c=13.397…, where the driving force for grain growth reverses its sign. This finding is also a purely topological characteristic of the polycrystalline network, one which does not depend on the kinetic rate-controlling mechanism.
- The independent partition of the total volume rate of change into its topological and metric components showed in all three cases a similar behavior. The metric component remains positive whereas the topological component remains negative, independent of the rate-controlling mechanism.
- Therefore, in spite of detailed quantitative differences, the overall behavior of 3-D network grains during self-similar growth remained unchanged for different *kinetic* controlling mechanism, namely, GB-, TJ-, or QP-mobility.

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