Limit conditions for the onset of abnormal grain growth in a homogeneous microstructure: Theory and Experiments

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Abstract. In previous papers it has been demonstrated that abnormal grain growth in a polycrystalline microstructure is a natural phenomenon even in the presence of an homogeneous microstructure. Namely, in a statistical sense, each portion of the material has the same grain size distribution and no local heterogeneities are involved. The only necessary condition is the presence of a grain growth inhibitor which can be represented by second phase particles (Zener Drag), homogeneously distributed as well, or by segregating atoms on the grain boundary (Atoms drag).

Considering specifically the Zener Drag effect, it can be shown that in the presence of an evolution of the restraining force (even if extremely slow), due to Ostwald ripening or to dissolution of the particles, the corresponding grain growth process allows the grain size distribution to continuously evolve reaching a critical shape. Such peculiar shape corresponds to a “structural” instability which irreversibly determines the onset of an abnormal growth phase with a drastic change of grain growth kinetics (kinetics exponents for the mean radius largely above the canonical 0.5).

In this context the mechanism of Abnormal Grain Growth onset is discussed as a general physical phenomenon and a set of experimental data on several steels will be presented for supporting such conclusion.

1. Introduction
The presence of second phase particles or solute atoms segregating along the grain boundaries affects the kinetics of grain boundaries motion resulting in a restraining force ($P_{Z,S}$) which reduces the kinetics of normal grain growth (mean radius $R=k t^n$ with $n<0.5$) [1-4].

Particles cause an inhibition of grain boundary motion which can be considered similar to a retarding force $P_Z$ acting along the moving boundary; there are thus two possible conditions:

- The external driving force $P$ has the same magnitude as $P_Z$ but opposite sign (the net force on the boundary is zero).
- The external driving force $P$ is greater than the maximum force $P_{Z0}$, which can be exerted by the particles, than the boundary can move.

The maximum force $P_{Z0}$ can be expressed as follows ($\gamma$ = boundary energy):

$$P_{Z0} = \gamma \cdot I_{Z0} \quad ; \quad I_{Z0} = \beta \cdot \frac{f_v}{r_p},$$

with $f_v$ (volume fraction of particles), $r_p$ (mean radius) and $\beta$ a constant in the range of 0.75 ÷ 2 according to particle geometry. The grain boundary velocity can be calculated as follows ($m$ = boundary mobility):

$$V = \frac{1}{m} \cdot \gamma \cdot I_{Z0} \quad ; \quad I_{Z0} = \beta \cdot \frac{f_v}{r_p}.$$
\[ v = m \cdot \Delta P = m \cdot (P - P_{z0}) \]  \hspace{1cm} (2)

In figure 1 the net forces \( \Delta P \) according to Eq.2 are plotted as a function of boundary velocity pointing out three ranges for such a function [2,3].

If we apply Eq. 1 and 2 to the ranges described above we obtain the values of \( \Delta P_{ij} \) as a function of the different boundary curvatures \((R_i, R_j)\):

\[
\Delta P_{ij}(I) = \gamma \cdot \left( \frac{1}{R_j} \right) \left( R_i - I_{z0} \right) ; \hspace{0.5cm} \Delta P_{ij}(II) = 0 ; \hspace{0.5cm} \Delta P_{ij}(III) = \gamma \cdot \left( \frac{1}{R_j} \right) \left( R_i + I_{z0} \right)
\]

(3)

Under the homogeneity assumption valid in any statistical system [2-4] it is possible to use the following kinetics equation for grain \( i \), by summing the motion of \( i \) grain boundary subjected to the net driving force \( \Delta P_{ij} \) and moving with \( v_{ij} \) velocity

\[
\frac{dR_i}{dt} = \sum_j \Delta P_{ij} = \sum_j w_{ij} v_{ij} = M \sum_j w_{ij} \left( \frac{1}{R_j} - \frac{1}{R_i} \right) ; \hspace{0.5cm} w_{ij} = \frac{\varphi_j R_j^2}{\sum \varphi_j R_j^2}
\]

(4)

where the expression for the probability function \( w_{ij} \) is derived from basic topological laws in [4] and \( M = 2m \gamma \) is the boundary diffusivity.

The growth rate for each class \( i \) can be derived specializing Eq.4, according to Eq.3, as :

\[
\frac{dR_i}{dt} = M \left[ \psi_I \cdot \left( \frac{1}{R_c} - \frac{1}{R_i} \right) + \psi_{II} \cdot \left( \frac{1}{R_c} - \frac{1}{R_i} \right) - \psi_{III} \cdot \left( \psi_I - \psi_{III} \right) \cdot I_{z0} \right]
\]

(5)

with \( \psi_I = \frac{\sum \varphi_j R_j^2}{\varphi_j R_j} ; \hspace{0.5cm} \frac{1}{R_c} = \frac{\sum \varphi_j R_j^2}{\sum \varphi_j R_j} \)

(6)

and \( \lambda = I, II, III \) interval [3]. By analyzing, in respect to a grain \( R_i \), the GSD we can divide it into three zones, as pictured in figure 2: one containing the grains which tend to shrink, one containing the grains which tend to stagnate and the last one containing the bigger grains which are allowed to grow when in contact with the grain \( R_i \). After the initial period of restricted grain growth, in the presence of stable grain growth inhibition, the mean radius tends to stabilize around a constant value [1-3]. It has also been shown that, starting from stagnation condition, for a decreasing value of the Inhibition factor \( I \), the mean radius tends to reach a stagnation condition \((n=0)\) characterized by a GSD, converging to a very well defined shape, independent of the initial conditions and the characteristics of which are [2]:

**Figure 1.** GB velocity vs. driving force \( \Delta P \) in the presence of Zener drag.

**Figure 2.** Grain Size Distribution (GSD) and relative behavior in correspondence to grain \( R_i \).
A GSD characterized by the absence of small grains, which disappeared during the restrained grain growth process, up to a value \( u_{\min} = R_{\min}/R_c \) which is in equilibrium with the largest grains in the system \( u_{\max} = R_{\max}/R_c \) (\( R_c \) is the critical radius in the classical sense and calculated in other papers [1,2]).

A peaked GSD (density function) with a maximum in correspondence of the smallest grains present in the system \( (R_{\max}/R_{\min} = 3, p(u_{\min})/p(u_{\max}) = 3^{1/4}, p(u) = (12/13) * 1/ u^{4}) \).

The stability analysis of the limit GSD at any, even infinitesimal, drop of the Zener Drag show that in this case the boundary energy minimization process reaches maximum instability [2].

Such approach to a limit GSD as a precondition for the onset of abnormal grain growth (AGG) can be more clearly appreciated in the presence of homogeneous Zener Drag instability.

In figure 3, in a double logarithmic plot, the case of normal grain growth \((l_z=0)\), starting from a log-normal distribution with mean radius of 5 micron, is shown. In this case the kinetics exponent of the mean radius is \( n=0.5 \) and the steady state distribution is the Hillert type for 3-D case [3].

Moreover in figure 3 a set of simulations are also shown where the Inhibition factor \( I_z \) is allowed to decrease continuously with a simple linear function but with different kinetics parameters \((dl_z/dt)\) according to the law \( I_z = I_{zo} - (dl_z/dt) \ t \) with \( I_{zo} = 1000 \ cm^{-1} \).

The effects, although of the same nature, are different in intensity according to the kinetics of the allowed inhibition drop. Namely, through a faster growth of the mean radius, larger abnormal grain sizes are obtained the slower the inhibition drop kinetics is (see the maximum coefficients \( n \) in figure 3; the apparent grain growth exponents for the mean radius are much higher than 1/2 while the maximum radius grows with \( n=1 \) according to the physics of the phenomena). After the fast growth of the mean radius (size of millimeters can even be reached, as it is well known in practice), proceeding the decrease of \( I_z \) until zero, the growth process converges to the normal grain growth “route”. However at this point, the growth process proceeds with much slower kinetics after the dramatic and fast change of grain size (and correspondingly to the change of the total boundary energy in the system). If the Inhibition drop is arrested during or after the discontinuous process of grain growth the system will reach a new stagnation condition, finding a new equilibrium between the value of the residual Inhibition and the grain size reached by the abnormal growth (even in this case the final result will be strongly influenced by the drop rate of the Inhibition value during the unstable period).

![Figure 3. Mean radius evolution as a function of different GG Inhibition drop rate.](image)

In figure 4, starting from the B GSD of figure 3, the evolution of the GSD is shown for a select case of Inhibition drop kinetics. Here it is possible to see how the instability in the Zener Drag
restraining force, through a continuous change of the GSD, leads to an unstable GSD which presents a high peak concentrated around relatively small grains in the system. For any further drop of the inhibition intensity this unstable condition develops in fast grain growth kinetics, characterized by a broader GSD (even double peaked if the drop rate is sufficiently slow) and ending up with large average grain size (from the initial grain radius of a few microns to the final one in the order of a millimetre). When the inhibition fades \((I_z \rightarrow 0)\), a steady state grain growth tends to establish\((t_s\) case in figure 6) which is asymptotically approaching the theoretical “route” for the normal grain growth shown in figure 3 \((n\) coefficient = 0.5). In figure 5 and 6 the shape of the grain size density function is compared at different stages of the process.

The evolution of the GSD suggests an “apparent” strategy adopted by the grain system to efficiently reduce the free energy (grain boundary energy) in the presence of a restraining force to a free grain growth (here specifically of Zener Drag type, but similar behavior can be expected also for the atoms drag [3]).

![Figure 4](image)

Figure 4. Evolution of GSD during Inhibition drop at different stage of the process (case B of figure 3).

In fact the change of the shape of the GSD, as previously described, suggests a kind of “preparation” of special topological conditions for the larger grains, namely the realization of a surrounding grain neighborhood formed preferably of the smallest grains available in the system, producing an energetically “unstable” equilibrium condition. For any further, even little, variation of the inhibition intensity the few relatively large grains will grow at the expenses of the smallest grains in the system with the most efficient kinetics \([4,5]\). In figure 7 the variation coefficient and the ratio between maximum and mean radius (of the simulated cases in figure 3) are shown as a function of time. It can be seen that the initial narrowing of the distributions is followed by a peak which becomes higher the slower the inhibition drop rate is during the abnormal process (coincident with max \(n\) values in figure 3) but never overtakes the theoretical limit value of \(p(u_{\text{min}}) = 4.67\) \([2]\). As a matter of fact, the limit distribution can never be exactly reached during the grain growth process if the presence of a certain thermally activated decay of the restraining force is present (particles dissolution or Ostwald ripening, as normally is the case). Moreover, such distribution should be seen as the final stage by which the microstructure loses “memory” of the initial GSD completely and therefore its shape becomes an invariant to any “path” the initial microstructure goes through.

2. Comparison between experiments and theoretical predictions

2.1. Structural Carbon Steel
Experiments on a set of Cr-Mn micro-alloyed steel, for quenching and tempering, either alloyed with Ti or Nb, have been carried out in laboratory. After ingot casting and hot rolling, samples of steel plates have been prepared to perform thermal treatments to study austenitic grain growth. The steels behave very similarly although the typical critical temperature of AGG changes with the chemical composition. For sake of brevity, only a Nb alloyed steel is hereby considered, with the following chemical composition (wt %):

|   | C   | Mn  | Cr  | Alsol | Nb  | N   | Si  | S   | P   | Ti  |
|---|-----|-----|-----|-------|-----|-----|-----|-----|-----|-----|
|   | 0.7 | 1.28| 0.52| 0.031 | 0.088| 0.012| 0.3 | 0.01| 0.02| _   |

The thermal treatments (1050 °C, for 8, 16 and 24 hours) are carried out in a laboratory furnace with controlled atmosphere. Grain boundaries, after martensitic quenching and tempering, are evidenced by a picric etchant with special additives, capable to reveal the prior Austenite grains. Grain size has been obtained by measuring the area of about 1000 grains for each sample by using a quantitative metallographic system. From the grain area an equivalent radius (diameter) of each grain can be defined and the 2-D grain size distribution so obtained has been transformed in 3-D by numerical procedure using the Schwarz-Saltykov method [7]. With this method the approximation of a spherical shape for the grains can induce to an overestimation of small grains in 3D, which requires a correction, typically involving less than 5% of total grain population. An analysis of the statistical significance of the histogram, by the numerosity of each grain class and in relation with the total number of the grain population, led to the choice of a size class width (radius) for the histogram equal to 2 micron.

Because of this procedure a series of possible sources of errors are concurrently operating and must be counteracted as much as possible by adopting accurate procedures, for instance:

- creating a sufficient homogeneous material along the steel fabrication
- adequately singling out the stage at which abnormal grain growth starts (Temperature and Time)
- controlling the quality of micrographs obtainable to measure the prior austenitic grain (quality of etchant, possibly needed correction for evident incomplete boundary patterns, use of a magnetic table to contour the grains,...)

- adopting a sufficient numerosity of the sample and good quality of transformation from 2-D to 3-D

In figure 5 the size distributions, relative to the three different treatment times, are reported and compared (grain frequency in % vs diameter in μm). It is possible to clearly see that there is an evolution of the shape of the size distribution and of other average parameters (figures. 7a,b,c).
Such a behavior is in line with the previous discussion of having an initial distribution narrowing in the presence of strong inhibitors of grain growth by the disappearance of very small grains in GSD (8h GSD in figure 5a) [1]. During the coarsening of (NbC,N) precipitates (continuous inhibition drop), the GSD evolves into a more peaked distribution (16h GSD), with an even narrower shape (see the variation coefficient in figure 5b), a ratio between maximum and critical radius around 2,1) and a ratio between the Mean radius and critical radius which reaches a value near the theoretical one for the limit distribution (12/13=0,923, figure 7a). The process is then followed by an initial phase of abnormal growth (24h GSD) in which a certain number of very large grains (5-10% in volume) appear in the microstructure. This can be statistically “measured” by: a shift of the peak of the 16h GSD to the left (smaller grains), a smaller ratio between the Mean radius and Critical radius - after having reached a value near the theoretical one for the limit distribution shown in figure 7a, and a variation coefficient that increases (also in correspondence of the creation of a tail of large grains in the GSD). The whole process is described in the simulation reported in figure 7d (peaking, shifting and tail creation in GSD) which corresponds to the evolution of distributions in first part of figure 4 up to the onset of AGG.

Figure 7a. Comparison as a function of time (h), between the experimental $R_{\text{med}}/R_{\text{crit}}$ ratio with the limit value 0,923.

Figure 7b. Comparison of the experimental values for the variation coefficient $k$ with the limit value: $k = 0.288 = 1/\sqrt{12}$.

Figure 7c. Evolution of $R_{\text{med}}$ and $R_{\text{crit}}$ parameters as a function of time. First converging and then diverging. It means that the variation coefficient first is pointing toward the limit condition $k=0,288$ , narrowing the GSD and then, after the onset of AGG, starts to increase.

Figure 7d. Evolution of the GSD in the simulation of Figure 3-case B, showing the approaching to the limit GSD and the deviation as soon the AGG starts (see GSD at $t_4$).

2.2. Grain Oriented Electrical Steel

Two grades of Conventional Grain Oriented Electrical Steels of the following composition (wt%) have been investigated for grain growth process up to abnormal growth condition.
Steel 1 contains only MnS, CuS precipitates and Steel 2 almost only contains AlN as second phase grain growth inhibitors (the latter by adding Aluminum and reducing the slab reheating temperature to a value in which the dissolution of Manganese Sulfide is negligible). The outcome of the growth process in these materials (Goss grains), is also influenced by the crystallographic texture in the primary matrix and is sensible to differences in the microstructure along the sheet thickness. However, the general grain growth process, before the establishment of the selective growth of the minor Goss Texture Component, follows with good approximation the rules of uniform grain growth in the presence of Zener Drag. The particle dissolution for Ostwald Ripening then induces microstructure evolution and instability.

Samples (30mm wide, 300 mm long // RD) from a cold rolled and decarburized strip (0.30 mm thick) have been treated in a special Thermal Gradient Furnace to simulate the box annealing treatment of secondary recrystallization (30°C/h up to 920°C, as maximum temperature on one end of the long side of the sample, with soaking treatment of 1 hour). Such treatment allows to single out the position (and the associated soaking Temperature) where the secondary recrystallization starts along the sample (in this experiment it was about 875 °C). It means that on the side of the sample above this temperature the secondary recrystallization is fully accomplished (secondary grains of millimeters size are visible to the naked eye) whereas on the side where the temperature is below 875 °C the grains are, in the stage of incipient AGG, continuously growing (typically in the range of 10-20 micron size). A metallographic sample just in front of the position in which the abnormal grains were visible has been taken (in the center of sheet thickness to avoid subsurface layer effects), namely where the continuous grain growth process was still operating, and then investigated for the GSD by measuring the area of more than 1000 grains. The 2-D grain size distribution has been transformed in 3-D by the method already mentioned [7]. The resulting experimental GSD shown in figure 8a refers to the case of Steel 1 at 870°C, which is very close to the temperature corresponding to the large secondary grains in the strip (875 °C in the temperature gradient). 875 °C here represents the case of onset of abnormal growth, in fact it’s possible to see in the distribution the appearance of a tail of large grains and correspondingly the presence of a class of small grains. The difference between the two histograms, in figure 8b, shows this process even better at the two ends of the GSDs, as well as the variation in the body of the histograms, with the increase of presence of grains roughly above the conventional critical radius (9,51μm), and a decrease of grains just below the R_c. The coincidence of the conventional critical radius valid for normal grain growth R_c is valid, as a parameter for the whole distribution, only for the limit distribution[2]. The equations (5) and (6) are those that regulate the growth in the general case, with the consequence of the fall of the concept of a unique critical radius valid for the whole distribution. The difference in the shape of the two GSDs (figure 8a) is again of the type described by the simulations in the figure 7d, namely the GSD in t_1 represents the sample at 875°C, with the typical shift in respect to the one near the critical distribution (sample at 870 °C). In figure 9a the GSDs of Steel 1 and 2 at 870°C (namely just before the onset of the secondary recrystallization) are compared. The important fact shown here is that notwithstanding the grain growth inhibitors being completely different in the two cases (AlN and (Mn,Cu)S), the GSDs are substantially identical showing again that the process is rather generalized and does not depend on the nature of the material or on the type of precipitates (see also the case of structural steel previously discussed). In figure 9b the comparison of Steel 2 at 870 °C with the limit distribution, transformed in a frequency histogram with the same class width and normalized with the same experimental critical radius, shows a very good agreement.

The above results and other similar ones for silicon iron already reported [3] support the evidence that the onset of abnormal grain growth here is preceded by a transformation of the GSD providing the topological condition for a fast discharge of the grain boundary energy.
3. Conclusion

In the framework of the Statistical Theory it has been shown that the abnormal grain growth in the presence of unstable inhibitors (Zener or Atom Drag, ..) is a continuous and homogeneous process and it is independent of local phenomena (as those possibly induced by topological, texture, precipitation or drag inhomogeneity) . The main characteristic of the grain growth process in these conditions is that the evolution of the GSD, according to the specific inhibition drop rate, passes close to a limit equilibrium GSD which, in the presence of a restraining force to grain growth, plays the role of “microstructure organization for efficiently reducing the boundary energy”. The formation of a microstructure with very few grains of relatively large size (3 times the smallest grains in the system) embedded in a large population of small grains (coincident with a high peak in GSD) produces an acceleration phase for the grain growth kinetics with features of a discontinuous process. The decay
rate of the inhibition is responsible for the growth kinetics; in fact a lower drop rate increases the slope of the mean radius kinetics, enhances the presence of a two peaks distribution in correspondence of the maximum growth speed and enlarges the size of the final abnormal grains. The comparison of the experimental GSD of steels just before the abnormal grain growth induced by Zener drag shows a good agreement with the theoretical prediction of the limit GSD (in particular with the samples of Micro-alloyed steels for quenching and tempering and of Grain Oriented silicon iron).

Accurate grain size measurements on GO Silicon Iron (Ferritic Phase) and Construction Steels (Austenitic Phase) show that this basic mechanism is very generalized and, in substance, is dominating any other possible growth enhancing effect in the microstructure (like for example texture effect in Grain Oriented steel for which the controlled drag evolution becomes a “necessary condition” for a proper texture selection).

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