

Nucleation and Growth During Recrystallization

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The evolution in the understanding of the recrystallization phenomena is summarized in this paper. Initially the main developments concerning recrystallization are presented from a historical perspective. Definitions and concepts involving recrystallization are presented regarding it as a solid-state reaction that occurs by nucleation and growth. The recrystallization nucleation mechanisms are subsequently discussed. Finally, the growth step is highlighted, emphasizing boundary and sub-boundary mobilities and the forces acting on the high angle grain boundaries that sweep the microstructure during recrystallization.

Keywords: *recrystallization, recrystallisation, recovery, nucleation, growth*

1. Historical Background

The production of metallic materials is impressive not only due to the quantity produced, more than 10⁹ tons/year, but also due to its diversity. It is estimated that there are more than 40.000 metallic alloys. Forging, rolling, extrusion and drawing deform about 90% of the production of metals and alloys. The knowledge of the phenomena such as work hardening, recovery, recrystallization, and grain growth is of fundamental importance not only to correctly work these materials but also to control their microstructures and optimizing their properties.

The plastic working of metallic materials has been practiced for millennia. For instance, people that lived five thousand years ago in the regions next to the Persian Gulf, Irak nowadays, have mechanically worked copper, gold, and silver. Therefore, some knowledge concerning strain hardening, softening by annealing and by hot working was already available at that time. The knowledge available of these phenomena, at the time of the great discoveries, are well summarized in Biringuccio's quote in 1540 in the classical book *De La Pirrotechnia*:

"Every work of gold and silver as well as copper may be worked either hot or cold, only you must be careful to anneal the thing at every hammering or when it needs it"

Kalisher¹, in 1881, in Germany, working with zinc strips, suggested that plastic deformation "destroyed the crystallinity" of the zinc, however subsequent annealing caused its restoration. It was then that the misleading term "recrystallization" has been given birth:

"Der Vorgang kann demnach so gedacht werden, dass das Zink seine krystallinische Struktur, die es beim Erstarren annimmt, durch das Walzen verliert und wenn es auf eine Temperatur erwärmt wird, welche diejenige, die es beim Walzen erhaelt, überschreitet, die krystallinische Struktur wiedergewinnt."

Usage of the optical microscope to observe steel microstructure, i.e. the invention of metallography by Sorby^{2,3}, perhaps may have been the most important fact in the XIXth century, employed in studies of recrystallization. Later, Stead⁴ affirms that recrystallization occurs by nucleation and growth, an important step forward to understand this solid state reaction. Ewing and Rosenhain^{5,7}, in works

published in 1899 and 1900, mention that polygons (polyhedra, in three dimensions) that appear in the polished and etched surface of metallographic samples are crystals. Amongst other things, they conclude that metals deform plastically through slip of their crystal planes and that they do not loose crystallinity during deformation. However, only in 1911/12, von Laue⁸ discovered the diffraction of X-rays on crystals, a powerful technique later used to confirm the crystallinity of metals. At the time of this experimental progress, crystallography was well established theoretically. For example, the book of Miller (*A treatise on crystallography*) edited in Cambridge in 1839⁹, has found fertile soil to develop and disseminate with aid of X-ray diffraction. In 1914, appeared the first book¹⁰ on this new area of knowledge, named *physical metallurgy* that later in the 60's would be transformed into *materials science*.

Recrystallization studies started and in 1920's the first reviews^{11,12} and the volume of results obtained and their importance were such that these phenomena were treated as a book chapter¹³. At the beginning of this decade, Alterthum¹⁴ published a work, which was conceptually very important, in which he affirms that recrystallization and grain growth have distinctive driving forces.

In 1924, Wever¹⁵ published the first pole figures on cold rolled aluminum and iron. In 1931, Burgers and Louwense¹⁶ have suggested that the recrystallization texture of aluminum single crystals was a result of oriented nucleation ("oriented nucleation theory"). The "oriented growth theory" only gained strength at the beginning of the 1950's, defended by Beck and collaborators¹⁷.

In 1934, Orowan¹⁸, Taylor¹⁹ and Polanyi²⁰ presented, independently, the dislocation concept. The way was opened for the understanding of work hardening and the phenomena that occurred during annealing of a work hardened material, however, the experimental tools to get irrefutable microstructural evidences were still lacking.

Kinetic recrystallization studies were carried out nearly independently from the studies on recrystallization mechanisms. In 1940, it was already known that the recrystallization kinetics (recrystallized volume fraction as a function of time, for isothermal annealing conditions) followed the KJMA relationship (Kolmogorov²¹ – John-

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son-Mehl²² – Avrami^{23–25}). In 1941, the book of W. G. Burgers²⁶ was published, probably the first book on recrystallization, a classical reference for many decades.

In 1949, R. W. Cahn²⁷ published in the *Journal of the Institute of Metals, as a Student Member*, his classic work on *polygonization* during annealing of single crystals deformed plastically by bending. This work was an important milestone since it used in a pioneering manner the dislocation concepts and presented experimental evidence obtained by optical microscopy of the dislocation rearrangements during annealing of worked crystalline materials. Heidenreich²⁸, also in 1949, observed for the first time, with the help of a transmission electron microscope (TEM), subgrains in aluminum. From that time the models of “recrystallization nucleation” by subgrain growth, proposed by Beck²⁹ and Cahn³⁰, and by strain induced migration of grain boundaries, proposed by Beck and Sperry³¹ were also introduced. Both models were suggested based upon observations made by optical microscopy.

Burke and Turnbull³², in 1952, summarized the knowledge available at the beginning of the 1950’s in a practical and objective way in the form of the so-called 7 “laws of recrystallization”. It is interesting to observe that the fundamental concepts such as *dislocation, nucleation and growth and crystallographic texture*, have not been mentioned in the recrystallization laws.

With the advent of transmission electron microscopy and further improvements on the dislocation theory, the deformation substructure and its evolution during annealing could be investigated in greater detail. At the beginning of the 1960’s, Swann³³ studied the dislocation arrangement of metals and alloys with a FCC structure. Keh e Weissmann³⁴ did the same for the BCC metals and alloys. In particular, important papers like those written by Hu^{35,36}, proposing recrystallization nucleation via subgrain coalescence by subgrain rotation and those from Bailey and Hirsch³⁷, observing in detail the migration of strain induced grain boundaries during annealing, must be listed. In both cases, transmission electron microscopy was essential to observe substructures and crystalline defects.

At the end of the 1950’s, Lücke and Detert³⁸ proposed the first quantitative theory on the interaction of solutes and moving grain boundaries. At the beginning of the 1960’s, Doherty and Martin³⁹ showed that precipitate dispersions can either accelerate (coarse dispersion; large dispersed precipitates) or retard (fine dispersion; small close precipitates) the recrystallization.

In the early 1970’s, a group of researchers of Sussex University, having R. W. Cahn and R. D. Doherty as leaders, started the series of experimental studies^{40–44} about the influence of deformation heterogeneities on recrystallization and subsequent modeling⁴⁵. Still in the 1970’s, Haeßner organized in Stuttgart a series of presentations on different aspects of recrystallization, given by a dozen of invited specialists. These presentations have been edited in a book form in 1971, whose second edition published in 1978⁴⁶ much contributed to the organizing of the knowledge and nomenclature of the area.

During the 80’s, two improvements in experimental techniques have been made available, more powerful for the study of recrystallization and related phenomena. One of them was the improvement of the techniques related to determine local orientation relationships (in the μm range) in crystalline materials, particularly the diffraction of backscattered electrons (“Electron Backscattered Diffraction – EBSD), utilizing the Kikuchi lines generated in the scanning electron microscope⁴⁷. With the help of EBSD it became possible to determine, with relative accuracy and speed, the orientation of a large number of micro-regions with dimensions of the order of about 0.3 μm or less, with the aid of field emission gun scanning electron microscopes (FEG-SEM). This technique allows obtaining the orientation distribution (macrotexture and microtexture) and the

boundary character distribution (mesotexture) in both deformed and recrystallized materials, as well as the detailed study of the type of grain boundaries^{48,49}. Another notable improvement occurred in the technique of determining the velocity (mobility) of boundaries and interfaces in solids⁵⁰. For example, nowadays grain boundary migration velocity in aluminum at high temperatures can be assessed in a continuous manner with accuracy in the range of 0.5 to 2%.

Finally, it should be mentioned that the great advance that occurred in the last years in the area of recrystallization seems to be due, once again, to the improvements in experimental techniques: the development of 3-D metallography with the use of high energy X-rays from synchrotron radiation beams for real-time *in situ* investigations⁵¹.

2. Definitions and Characteristics of Recrystallization

The microstructural changes occurring during annealing of a cold-worked metal lead to the decrease of the stored energy due to plastic deformation. This energy decrease is caused by mechanisms of rearrangement and annihilation of crystal defects. According to Haeßner⁵², the main changes are: 1) reaction of point defects leading to their decrease; 2) dislocation annihilation of opposite signs and shrinking of dislocation loops; 3) dislocation rearrangement in order to form lower energy configurations (e.g., low angle grain boundaries); 4) formation of high angle grain boundaries; 5) absorption of point defects and dislocations by migrating high angle grain boundaries; and 6) reduction in total grain boundary area. The microstructural modifications 1), 2), 3) and 4) are classified as recovery and the modifications 5) and 6) are classified as recrystallization and grain growth, respectively (Haeßner mentions only 5 processes). As the crystalline defects distribution after straining is in general heterogeneous, each material region can go through a sequence of microstructural changes as mentioned, at different times.

The “continuous” or “*in situ* recrystallization” terms have probably been used for the first time by Hornbogen⁵³ at the end of the 60’s for the case of aluminum alloys containing a dispersion of fine precipitates, in which recrystallization was inhibited but which softened considerably due to recovery (“extended recovery”). Until then, “continuous recrystallization” and “extended recovery” were considered synonymous. More recently, Humphreys and Chan⁵⁴ proposed to differentiate “extended recovery” from “continuous recrystallization” as a function of the relative quantity between high and low angle grain boundaries that were present in the microstructure. To that extent, the definition suggested by Haeßner and adopted in this text is in agreement with the proposal of Doherty⁵⁵:

“I would describe recrystallization as the change in grain structure of material by application of thermal annealing to samples that had been plastically deformed so as to eliminate almost all the dislocations introduced by the deformation by means of migration of high angle grain boundaries. This definition by including plastic deformation, removal of dislocations and migration of high angle grain boundaries covers almost all the major features of this process of changing the grain structure.”

Despite of the fact that recrystallization may be understood and treated as an irreversible, thermally activated solid state reaction which occurs by nucleation and growth, it presents numerous specific aspects that are worth mentioning:

- The driving force for recrystallization (stored energy due to deformation in the form of crystalline defects) is very small when compared to other solid-state reactions (see item 4) and is practically independent of recrystallization temperature.
- The classical theory of nucleation does not work well for recrystallization, because apart from the low driving force,

the interface energy between the recrystallized region and the non-recrystallized matrix (high angle grain boundary) is very high. As a consequence we have large nuclei, in the order of $\sim 1 \mu\text{m}$ or $\sim 10^{10}$ atoms, and it is difficult to imagine them as due to fluctuations.

- c) Recrystallization nucleation means formation and migration of large angle grain boundaries stemming from pre-existing embryos.
- d) There is experimental evidence of at least three nucleation mechanisms.
- e) The start (“nucleation”) occurs in an extremely heterogeneous manner, predominantly at deformation heterogeneities.
- f) When compared to other solid state reactions, a small number of nuclei per unit volume are formed. From each million of “embryos” (deformation cells or subgrains) only one of them transforms into a nucleus.
- g) After the initial nucleation stage only growth occurs, i.e., recrystallization is mostly a site-saturation solid state reaction.
- h) In general, growth velocity decreases with time. The reasons for this decrease in growth velocity with time are deformation gradients in the microstructure resulting from the deformation and competition with concurrent recovery.
- i) The resulting recrystallized grain size is very sensitive to the applied strain and less sensitive to the annealing temperature.

3. Nucleation or Start of Recrystallization

As we mentioned before, the term nucleation will be adopted in this text to indicate the beginning of recrystallization. The mechanisms of dislocation rearrangements in order to form a low dislocation density region associated with a large angle grain boundary with high mobility and therefore, capable of a fast migration over the strained (or recovered) matrix, will be defined as recrystallization nucleation.

It is unlikely that the classical homogeneous nucleation theory can be fully applied to recrystallization due to its low driving force unlike those ones associated with solidification or solid-state precipitation. Furthermore, grain boundary energies are much higher than those observed in other solid state reactions. In this regard, thermal fluctuations themselves cannot explain the formation of defect-free regions bounded by high angle grain boundaries upon annealing. Hence, the “nuclei” which give rise to the new recrystallized grains are not formed during annealing; they are already present in the de-

formed state. The deformation structures with high local orientation gradients constitute the pre-deformed nuclei. In this type of analysis, transition and shear bands are potential sites where these pre-nuclei may trigger recrystallization. The three best-known recrystallization nucleation models are described in the following.

3.1. Migration of pre-existing high angle boundaries

This model has been originally suggested by Beck e Sperry³¹ based upon observations made by optical microscopy and later studies with transmission electron microscopy by Bailey^{37,56,57} in Cu, Ni, Au, Ag and Al. In these studies, the metals were deformed less than 40%. This mechanism takes into account the migration of a pre-existing grain boundary toward the interior of a more highly strained grain, as shown in Figure 1.

The condition for the process to occur is the favorable energy-balance between the decrease of stored energy due to the elimination of defects caused by the passage of the boundary and the increase in total grain boundary surface due to bulging. Figure 2 illustrates the model schematically. The growth condition is given by:

$$L > \frac{2 \cdot \gamma}{\Delta E} \quad (1)$$

where γ is the grain boundary surface energy per unit area and ΔE is the released energy associated with the decrease in defects.

3.2. Nucleation by low angle boundary migration (sub-boundaries)

This model has been suggested independently by Beck²⁹ and Cahn³⁰ and with further improvements made by Cottrell⁵⁹ and is generally quoted in the literature as being the *Cahn-Cottrell model*. The model is based upon the polygonization phenomena where regions of low dislocation densities are surrounded by sub-boundaries. Once a subgrain is formed, it will be capable of growing at the expense of its neighbors by thermally assisted subgrain boundary migration. In this way, a decrease in stored energy will occur during annealing due the removal and rearrangement of microstructural defects. The moving sub-boundary absorbs dislocations, increasing its orientation difference, its energy and mobility until it is transformed into a high angle boundary, hence characterizing nucleation. Sandström and collaborators⁶⁰ observed the subgrain growth in pure aluminum at temperatures of 300 to 400 °C (0.6 and 0.7 T_m , respectively, were T_m is the absolute melting point), showing that this was the operative mechanism at higher temperatures. Varma and Willits⁶¹ observed

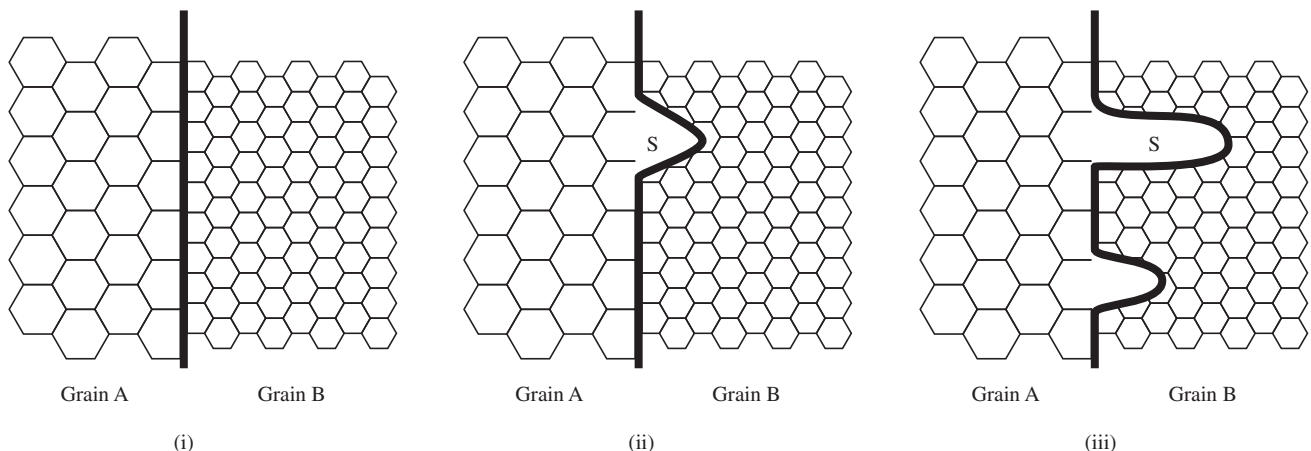


Figure 1. Schematic representation of nucleation by migration of boundaries induced by deformation⁵⁸.

essentially the same. Walter and Koch⁶² observed the mechanism of nucleation by migration of sub-boundaries associated with transition bands in a Fe-3%Si alloy using transmission electron microscopy. More recent experimental evidence on the migration of low angle boundaries during hot deformation was observed in pure aluminum and in Al-5%Mg alloy⁶³. Figure 3 illustrates schematically the mechanism of recrystallization start by sub-boundary migration.

3.3. Nucleation by subgrains coalescence

This model has been postulated in 1948 by C. S. Smith⁶⁴ and further studied on Fe-3%Si single crystals by Hu^{35,36}. Li⁶⁵ analyzed the process from the point of view of kinetics and thermodynamics

and concluded that it was a sluggish process. Later, Doherty and Szpunar⁶⁶ showed that coalescence of subgrains at temperatures lower than $0.65 T_m$ would be a kinetically viable process. Transmission electron microscopy observations carried out by Jones⁶⁷⁻⁶⁹ have shown the occurrence of the coalescence of two neighboring subgrains in aluminum. The mechanism was based upon the coalescence of two adjacent subgrains and which is equivalent to a rotation, causing the crystal lattices to coincide, as shown in Figure 4.

Still quoting the systematic study of Sandström and co-authors⁶⁰, subgrain coalescence has been observed in aluminum using transmission electron microscopy, for temperatures of 0.4 and $0.5 T_m$. Therefore, experimental evidences suggest that subgrain coalescence, when compared to subgrain migration, is favored for lower annealing temperatures.

Coalescence is a mechanism that promotes subgrain growth, disappearance of sub-boundaries (decreasing the stored energy) and changing the orientation differences between a group that went through coalescence and its neighboring subgrains. The increase in the orientation difference leads to the appearance of a high angle boundary capable of high-speed migration, forming the nucleus of recrystallization. It is important to point out that the global energy balance that takes into account the disappearance of sub-boundaries with the increase in orientation difference and the energy of others, is favorable, i.e., leads to a decrease in total free energy. Figure 5 illustrates schematically the formation of a nucleus by subgrain coalescence.

4. The Role of Deformation Heterogeneities on Nucleation

Plastic deformation rarely occurs in a homogeneous manner in most cases. In particular, single crystals of special orientations and coarse-grained specimens tend to display very heterogeneous deformation structures. In many cases, this heterogeneity is caused by the macroscopic subdivision of grains. Deformation heterogeneities like deformation and transition bands subdivide individual grains into distinct regions. Within each region, plastic deformation occurs in a quite homogeneous manner but different from the neighboring regions.

Deformation heterogeneities are regions that contain a larger quantity of crystalline defects per unit volume and present also larger

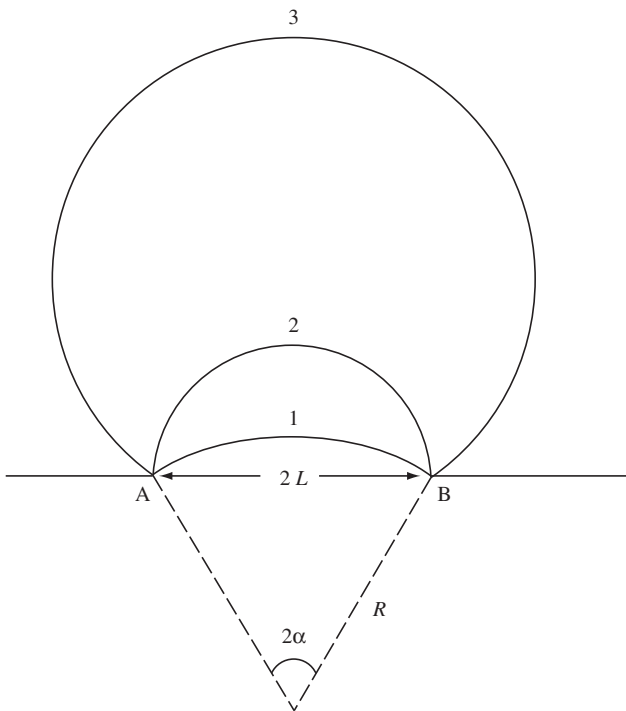


Figure 2. Model for the grain boundary migration induced by deformation, showing the successive migration stages 1, 2 and 3⁵⁷.

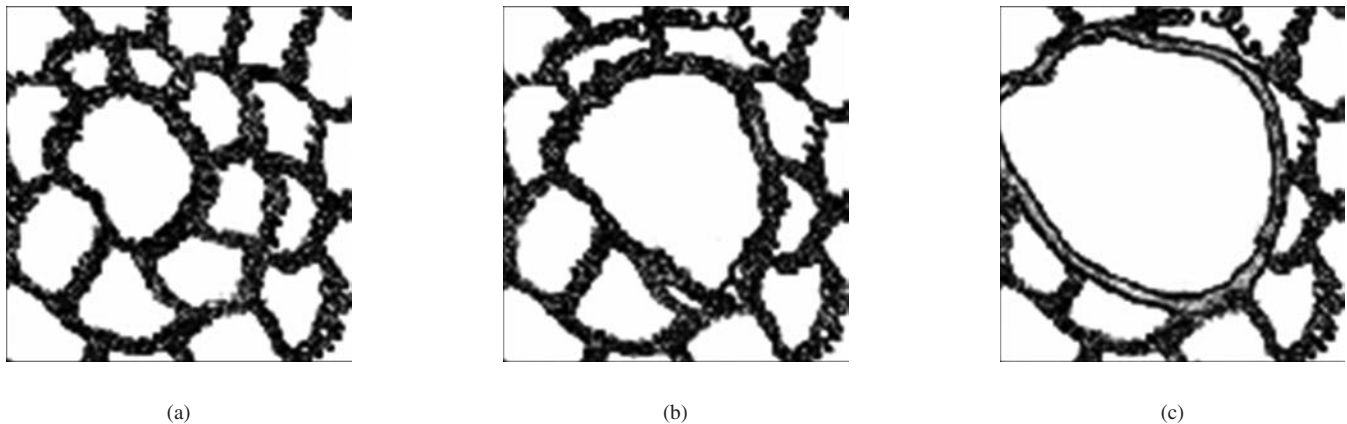


Figure 3. The sequence shows the nucleation of a recrystallized grain starting from a subgrain: a) initial substructure; b) the larger (middle) subgrain growth over the other (smaller) ones; and c) an area free of defects associated to a large angle boundary that is being formed.

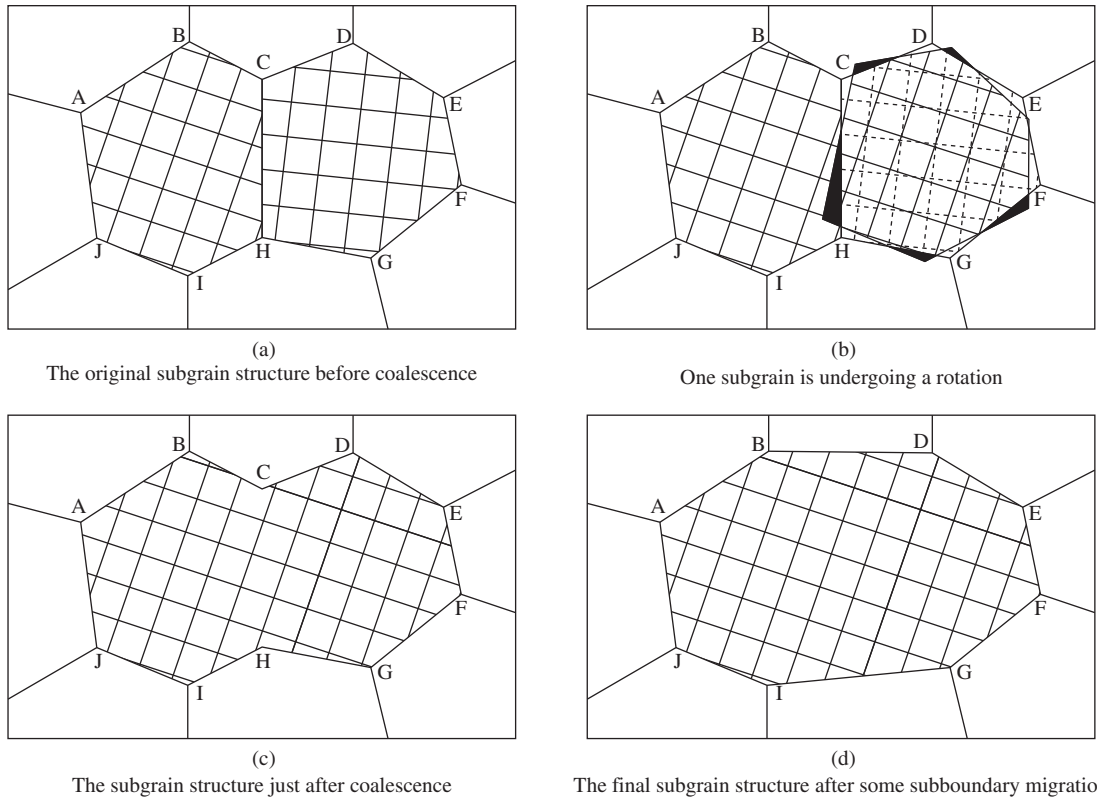


Figure 4. Coalescence of two subgrains by “rotation” of one of them: a) original structure prior to coalescence; b) rotation of the CDEFGH grain; c) subgrain structure subsequent to coalescence; and d) final structure after sub-boundaries migration⁶⁵.

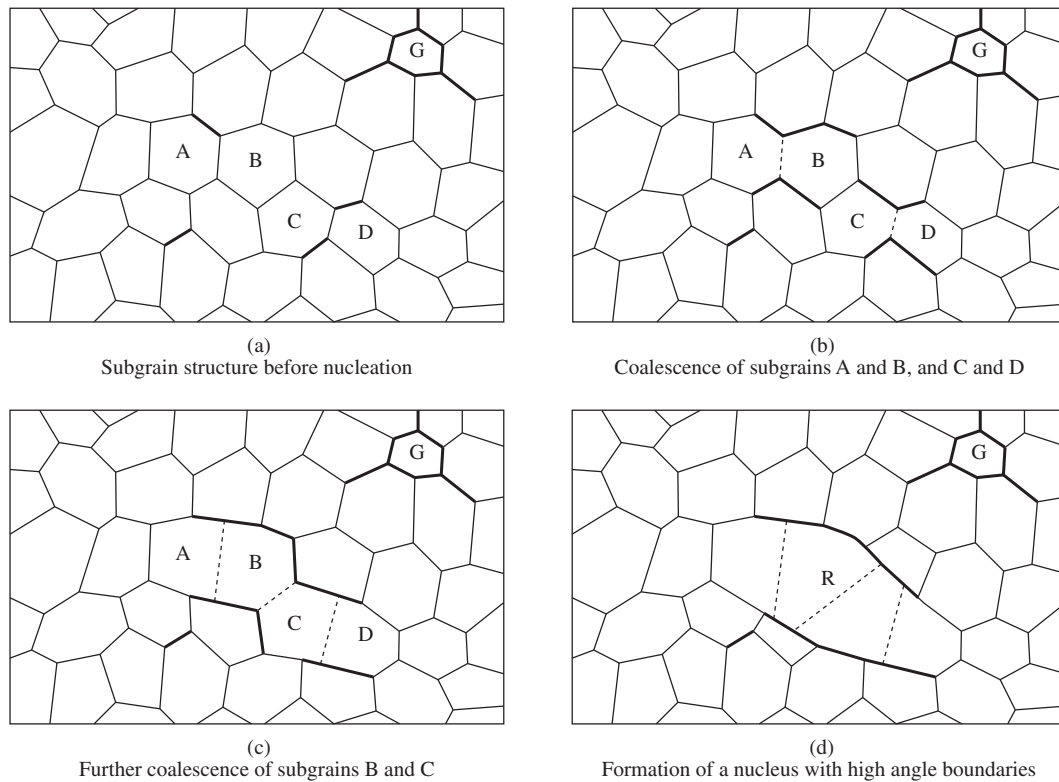


Figure 5. Schematic representation of a recrystallized grain. The orientation difference of subgrains is represented by the line thickness: a) original structure; b) coalescence of pairs by elimination of common boundaries; c) coalescence of the A/B and C/D pairs; and d) recrystallized grain originating from a group of coalesced subgrains. The thicker lines indicate large angle boundaries³⁶.

variation in lattice orientation (local lattice curvature). In a niobium bicrystal deformed by cold rolling to moderate strains, just to give a single example, highly misoriented boundaries were developed in the microstructure due to deformation banding, many reaching about 55° ⁷⁰. The number of defects is larger when compared with other regions of the plastically deformed metal, creating preferential sites for the occurrence of recrystallization nucleation. These regions contain “potential nuclei” or “embryos”, formed during plastic deformation. In metals where twinning, transition banding, and shear banding occur, nucleation will start initially near or at the intersections (in the case of twins) of these heterogeneities. The areas next to pre-existing grain boundaries, prior to deformation, are also preferential sites. Microscopic observation of samples partially recrystallized (with low volume fraction) reveals that the homogeneous distributions (or even nearly homogeneous) of recrystallization nuclei are very rare. Figure 6 shows an example of the recrystallization nucleation within shear bands in coarse-grained tantalum⁷¹. Note the predominance of potential nuclei (subgrains) within shear bands in comparison with the surrounding coarser subgrain structure.

5. Occurrence of the Three Nucleation Mechanisms

Considering the mechanism of migration of pre-existing grain boundaries, the basic requirement for it to occur is the existence of large strain differences between adjacent grains. Studies conducted by Beck and Sperry³¹ and Bailey^{37,56,57} showed that the mechanism at hand is important for deformations up to 40% reduction.

The largest dispute in the literature is associated with the occurrence of subgrain coalescence and migration of sub-boundaries. Ex-

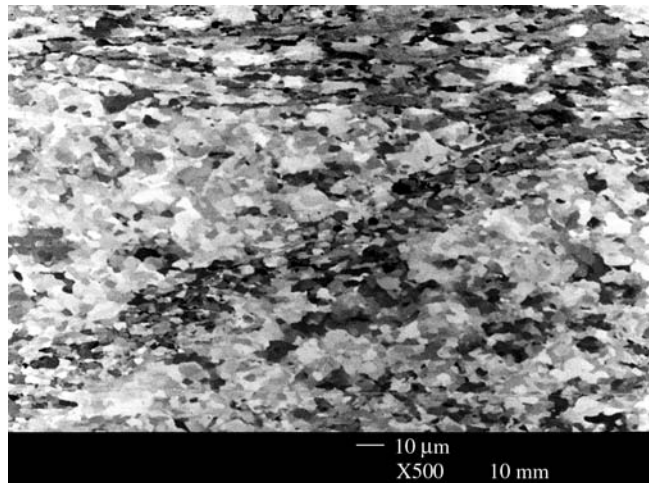


Figure 6. Heterogeneity in the distribution of recrystallized regions due to shear bands. SEM micrograph showing a sample of tantalum deformed by cold swaging to 72% area reduction and annealed at 900 °C for 30 minutes (BSE)⁷¹.

perimental data in the literature allows some preliminary prospective considerations to be made on the occurrence of the two last mentioned models. The subgrain migration mechanism seems to be associated with high strains⁷², large spread in the subgrain size distribution⁷³, relatively high annealing temperatures^{61,62} and occurs preferentially in low-SFE metals⁷⁴. The mechanism of subgrain coalescence, in turn, seems to be associated with transition bands⁷³, large spread in the distribution of subgrain angles^{73,75}, moderate strains⁷³, regions next to grain boundaries^{44,69,72}, relatively low annealing temperatures^{61,62,68}, and metals with high SFE⁷⁴. On the other hand, the evidences of the subgrain coalescence mechanism obtained from *in situ* observations on thin samples in TEM have cast some doubt if this mechanism is still applicable to thicker samples. Table 1 summarizes in a tentative way some conditions for which the different recrystallization mechanisms occur, as found in the literature.

During hot deformation, work hardening and restoration mechanisms occur simultaneously. Due to the high temperature, recovery processes take place at high rates, which decrease the driving force for recrystallization. In hot deformation, recrystallization is usually classified as “static”, which occurs after deformation or “dynamic”, which occurs during deformation. The occurrence of static or dynamic processes will depend on the applied strain (higher strains will favor dynamic processes), strain rates (lower strain rates will favor dynamic processes), stacking fault energy (higher energy will favor static processes) and the temperature. Among the nucleation mechanisms listed above, SIBM and its variations are most likely to take place during and after hot deformation.

In 1995, Nes⁷⁶ published a fairly extensive review on recovery, with a detailed mathematical treatment of the processes involved, including both mechanisms on recrystallization nucleation that take into account subgrain growth. The recent development of 3-D metallography⁵¹ allows the direct observation of the microstructure evolution in thick strained samples and may put an end to this dispute.

More recently, Ferry and Humphreys⁷⁷ have described the occurrence of discontinuous subgrain growth in deformed and annealed {110}<001>-oriented aluminum single crystals. After deformation the microstructure showed neither large orientation gradients nor heterogeneities, making the material resistant to recrystallization. Upon annealing, a small number of subgrains grew rapidly and discontinuously to diameters of over 100 μm. The authors attributed the discontinuous subgrain growth to orientation differences between the subgrains formed during deformation. Later, Monte Carlo simulations showed that abnormal subgrain growth is also a plausible mechanism for recrystallization⁷⁸.

6. Growth of the Recrystallized Regions

Migration of high angle boundaries is the fundamental mechanism acting during recrystallization and grain growth, the main difference between them being the driving force related to both phenomena. The driving force for grain growth, being it continuous (normal grain growth) or discontinuous (abnormal grain growth), is the energy of the high angle boundaries. The main driving force for recrystal-

Table 1. Tentative summary for the mechanisms describing the start of recrystallization.

Strain induced grain boundary migration	Sub-boundary migration	Subgrain coalescence
<ul style="list-style-type: none"> • small strains (up to ~ 40%) • hot working 	<ul style="list-style-type: none"> • high strains • high temperatures • heterogeneous subgrain size distribution • low-SFE metals and alloys 	<ul style="list-style-type: none"> • moderate strains • relatively low temperatures • large spread in the distribution of subgrain misorientations • transition bands • high-SFE metals and alloys

lization to occur is the stored energy during straining in the form of crystalline defects. In a region exempt of defects, surrounded by high angle boundaries, recrystallization proceeds by growth of this "nucleus" over the yet non-recrystallized matrix. Another important difference between recrystallization and grain growth is the curvature signal of the migrating high angle boundary. Figure 7 illustrates two examples of reaction fronts (high angle boundaries) advancing into the strained, or better, non-recrystallized matrix. In Figure 7a, the non-recrystallized region is a FCC solid solution of low stacking fault energy (SFE) that suffered little modification during annealing, whereas in Figure 7b, the non-recrystallized region of a pure BCC metal with a high SFE presented marked recovery.

Under isothermal conditions, primary recrystallization generally presents an incubation time, associated with the formation of a nucleus. The absence of the incubation time, generally is related to nucleation of the type of *pre-existing boundary migration*, not having in this case, the need of time for the formation of a high mobility boundary. Growth of the recrystallized regions due to high angle boundaries continues until mutual impingement of recrystallized grains. Primary recrystallization finishes when both reaction fronts meet. The distribution of the recrystallized regions is heterogeneous and grain impingement already occurs even for low volumetric recrystallized fractions.

The migration of high angle boundaries sweeping the microstructure and removing the crystalline defects is an irreversible process. When high angle boundaries move from left to the right, atom flux through the boundary from right to the left is higher than the flux in the opposite direction. Taking into consideration the model in which the atoms overcome individual and independently the activation barrier, by passing from the strained grains to the recrystallized grains, one gets for $\Delta P \ll kT$:

$$V = (A \cdot L \cdot f / kT) \cdot \exp(-\Delta A / kT) \Delta P \quad (2)$$

where V is the velocity of the interface (high angle boundaries, in the present case), ΔP is the driving force for the transformation, ΔA is the activation energy, A is an accommodation factor, L is the interface thickness, f is a characteristic frequency, T is the absolute temperature, and k is the Boltzmann constant.

Equation 2 shows that migration velocity of grain boundaries is proportional to the driving force for recrystallization and strongly depends on temperature. Migration velocity, V , is frequently presented in a simplified form as a function of two factors: boundary mobility, M , and transformation driving force, ΔP , and is given by the equation:

$$V = M \cdot \Delta P \quad (3)$$

It is important to mention that in the literature there are other formulations for the pre-exponential factor of Equation 2. In reality, the ΔP term is the summation of the driving forces and retarding forces acting on the boundary and will be discussed in greater detail in the following. Prior to that, however, boundary mobility will be discussed.

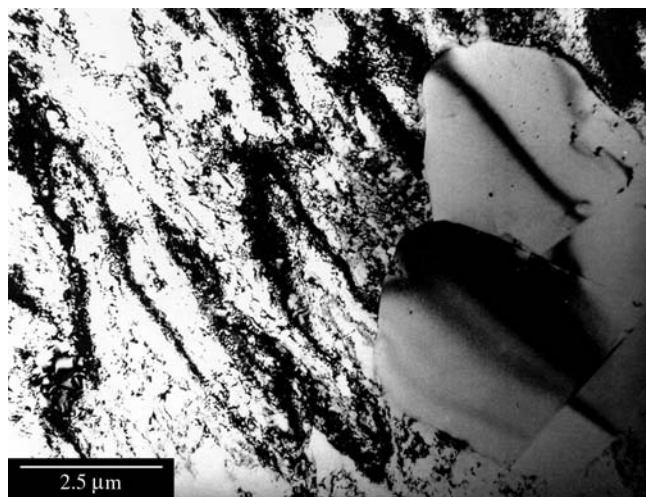
7. Boundary Mobility

We shall start this item with a phrase taken from the most recent review article⁷⁹ on boundary mobility:

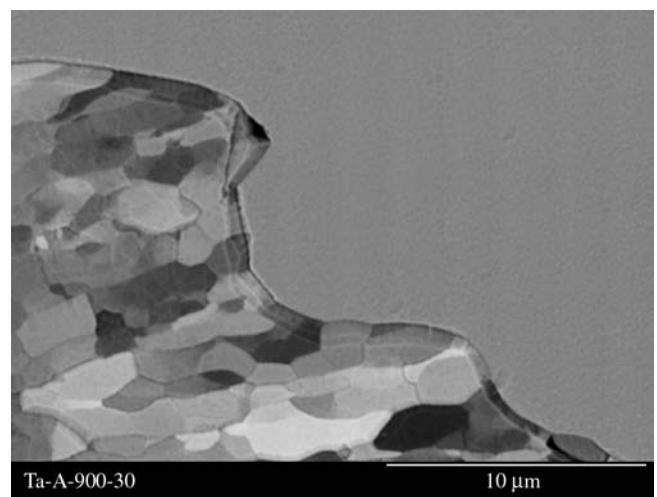
"Understanding the mobility of grain boundaries is a classic example of a problem in material science that is simple at the level of college student education but a significant challenge for those who would like to be able to understand it in detail."

Despite of this review⁷⁹ emphasizing the non-existence of a satisfactory theory related to boundaries, specially to high angle grain boundaries, nevertheless it is possible to evaluate the effect of some variables on the mobility. Equation 3, that describes migration velocity, does not leave explicit the influence of the crystallographic orientation, which has a considerable importance, known for more than 50 years. Figure 8, taken from Liebman's doctoral thesis presented in the 50's, shows that migration velocity varies considerably with the misorientation angle θ (difference in orientation between two grains).

Later works showed that amongst a large orientation variation presented by the boundaries, some distinct steps in mobility could be observed. For small differences in orientation of a few degrees (for example, subgrain boundaries), mobility is very small and can increase or decrease with the orientation difference for *mixed boundaries* or *pure tilt boundaries*, respectively⁸¹. Starting from orientation



(a)



(b)

Figure 7. Reaction fronts (high angle boundaries) migrating during recrystallization: a) transmission electron micrograph of an austenitic stainless steel containing 15% Cr and 15% Ni, cold worked to a 50% area reduction and annealed at 800 °C for 3 hours (by kindness of K. Ehrlich, Forschungszentrum Karlsruhe, FZK, Germany); and b) micrograph of a sample of pure tantalum cold worked to 72% area reduction and annealed at 900 °C for 30 minutes (SEM, BSE).

differences of about 1° , mobility increases with θ , as schematically shown in Figure 9. The high angle boundaries, with exception of the twin like boundaries, present a far greater mobility than low angle boundaries.

The activation energy for migration of low angle grain boundaries is very close to that one for volume diffusion (self-diffusion), suggesting that the process is controlled by the mechanism of dislocation climb. On the other hand, activation energy for large angle grain boundary is much smaller, about $3/5$ of the activation energy for migration of small angle grain boundaries, indicating that motion of these boundaries is controlled by the atom exchange through the moving boundary⁸². More recent work^{83,84} reviewing the influence of orientation difference on mobility, confirms that high angle boundary mobility ($> 15^\circ$) is about 100 to 1,000 times greater than the low angle

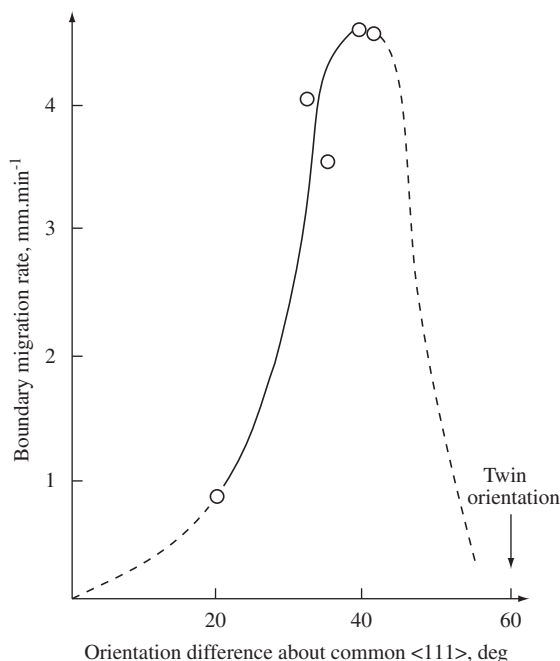


Figure 8. Change in migration rate of the grain boundary V in aluminum as a function of rotation angle in relation to $\langle 111 \rangle$ direction⁸⁰.

boundary mobility (2 to 5°). **Activation energies found in high purity copper** were 204 and 125 kJ/mol for low and high angle boundaries, respectively. These activation energies confirm the hypothesis that low angle boundary mobility is controlled by volume diffusion, while the boundary mobility of high angle boundaries is controlled by diffusion through the boundary. Recent experiments⁸⁵ with tricrystals of aluminum and zinc indicate that the sluggish movement of triple-point grain junctions may control boundary mobility. The geometry and configuration of the junctions play an important role in boundary mobility.

There is extensive evidence that mobilities and activation energies for migration of high angle boundaries are dependent on orientation⁸³. There are some characteristic orientations for which boundaries have an especially high mobility. Table 2 shows some examples of materials and their high mobility boundary orientations.

Although many unresolved questions remain about the effects of solutes on anisotropy, the currently available literature shows that significant anisotropy exists with respect misorientation angle, axis and boundary plane⁷⁹. Mobility of special boundaries is associated with the presence of impurities in solid solution (see Figure 10). These boundaries play an important role in secondary recrystallization, i.e.,

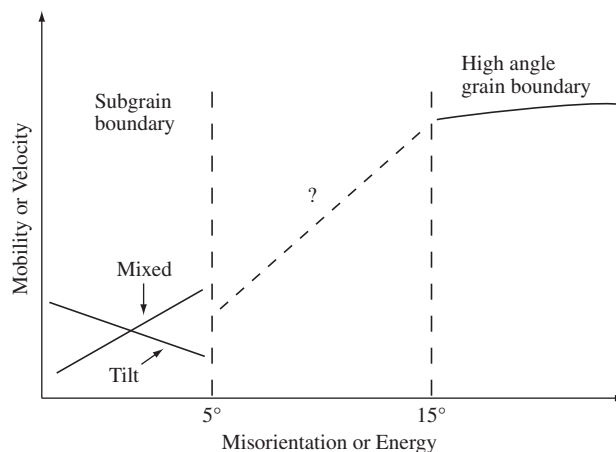


Figure 9. Changes in mobility as a function of the misorientation angle θ (schematic). Practically there are no data available in the literature for the range of $5^\circ < \theta < 15^\circ$.

Table 2. Crystallographic orientations for the occurrence of high mobility boundaries⁸³.

Nearest coincidence relationship			Experimental relationship		Metal	Structure
Sigma	Rotation	Axis	Rotation	Axis		
$\Sigma = 7$	38.2°	$\langle 111 \rangle$	$35-45^\circ$	$\langle 111 \rangle$	Al	fcc
			38°	$\langle 111 \rangle$	Cu	fcc
			$36-42^\circ$	$\langle 111 \rangle$	Pb	fcc
$\Sigma = 13a$	22.6°	$\langle 100 \rangle$	23°	$\langle 100 \rangle$	Al	fcc
			19°	$\langle 100 \rangle$	Cu	fcc
$\Sigma = 13b$	27.8°	$\langle 111 \rangle$	30°	$\langle 111 \rangle$	Cu	fcc
			30°	$\langle 111 \rangle$	Ag	fcc
			$20-30^\circ$	$\langle 111 \rangle$	Nb	bcc
$\Sigma = 13$	30°	$\langle 0001 \rangle$	30°	$\langle 0001 \rangle$	Zn	cph
			30°	$\langle 0001 \rangle$	Cd	cph
$\Sigma = 17$	28.1°	$\langle 100 \rangle$	$26-28^\circ$	$\langle 100 \rangle$	Pb	fcc
			30°	$\langle 100 \rangle$	Al	fcc
$\Sigma = 19$	26.5°	$\langle 110 \rangle$	27°	$\langle 110 \rangle$	Fe-Si	bcc

in situations where the driving force for boundary migration is much smaller⁸⁶. In a simplified and schematic manner, we might affirm (see Figure 10) that special boundaries present higher mobility and are less affected by atoms in solid solution^{87,88}. It is interesting to mention that coherent twins present the most coincidence sites ($\Sigma = 3$), yet they have almost zero mobility (see Figure 8).

Finally, it should be pointed out that mobility of mixed boundaries with small orientation differences (up to 5°) have been little studied in the literature. Regarding boundaries with intermediate orientations differences (from 5 to 15°), no study at all has been conducted. Therefore, it would be desirable that new studies should be conducted in these specific areas⁸⁹.

8. Driving and Retarding Forces

As discussed in the previous item, after the nucleus formation, recrystallization is controlled by growth, in which high angle grain boundaries migrate over the strained matrix (non-recrystallized), removing the crystalline defects until grains meet mutually. Elegant experiences with aluminum bicrystals⁹⁰ show in an unequivocal manner the influence of the driving force on the migration velocity of grain boundaries.

Grain boundary migration velocity during primary recrystallization (V) may be given (see Equation 3) by the following equation:

$$V = M \cdot \Sigma F \quad (4)$$

where $\Sigma F = \Delta P$ is the summation of the forces acting on the reaction front.

The grain boundary mobility has been discussed earlier. In this item we conduct an analysis on the forces that act on a high angle

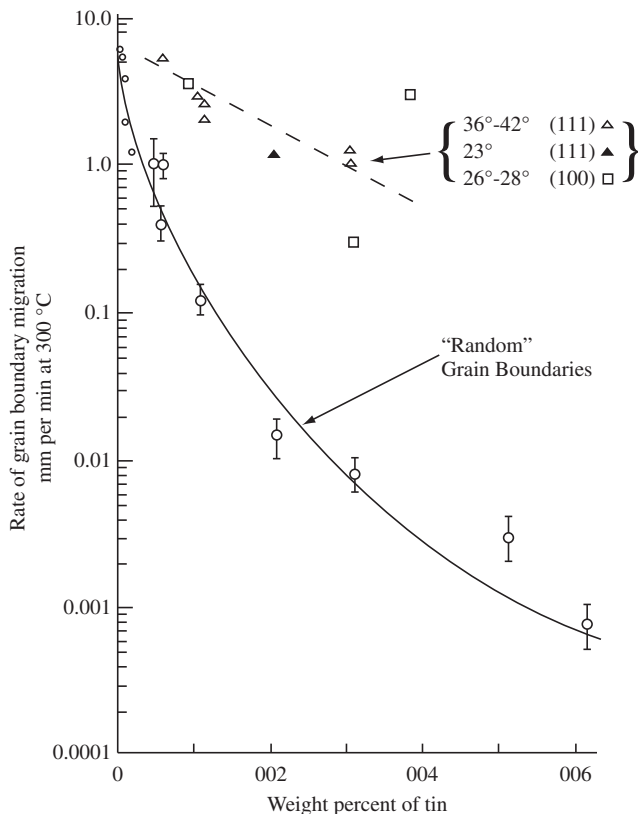


Figure 10. Changes on migration rate of grain boundaries of lead at 300°C as a function of tin content, for random grain boundaries and special grain boundaries^{87,88}.

boundary during its migration. The values of forces presented in the following represent a quantity of energy per unit volume of material, with $[\text{J}/\text{m}^3]$ units. However the $[\text{J}/\text{m}^3]$ unit may be represented by $[\text{N}/\text{m}^2]$ and by mechanical analogy, the acting potentials can be understood as “pressures” applied on high angle boundaries. According to literature^{53,91-93}, the main forces acting on the reaction front are: driving force due to straining, driving force due to grain boundaries, driving force due to discontinuous precipitation and retarding forces due to solute atoms and to precipitated particles. In the following we discuss in a separate manner these driving and retarding forces.

8.1. Driving force due to plastic deformation

During plastic working of polycrystals some important events occur:

- i) Grains change their shape;
- ii) Grains rotate and develop preferential orientations (texture);
- iii) The quantity (area) of grain boundaries per unit volume increases; and
- iv) The quantity of point defects and of dislocations per unit volume increases several orders of magnitude.

Part of the energy spent during the deformation process is stored in the material in the form of crystalline defects. The major contribution is due to dislocations. If the dislocation density prior to deformation is ρ_0 and ρ after deformation, the driving force F_N is given by:

$$F_N = G \cdot b^2 \cdot (\rho - \rho_0) \quad (5)$$

where G is the shear modulus and b is the modulus of the Burgers vector. In practice, the dislocation density after deformation is several orders of magnitude greater than the dislocation density of the material prior to deformation. In this manner, Equation 5 may be simplified to:

$$F_N = G \cdot b^2 \cdot \rho \quad (6)$$

For a material (copper, for example) with a dislocation density of $10^{11} \text{ cm}/\text{cm}^3$ in the strain hardened condition and of $10^6 \text{ cm}/\text{cm}^3$ after annealing, having $G \cdot b^2 = 10^{-8} \text{ N}$, we obtain $10^3 \text{ N}/\text{cm}^2$ for F_N . Table 3 compares driving forces (in J/mol) of several phase transformations. It may be observed that recrystallization is one of the solid state reactions with the smallest driving force.

8.2. Driving force due to the grain boundaries (grain growth)

The surface energy (γ) associated with grain boundaries with average diameter D is given by the relationship:

$$F_{gg} = (2\gamma / D) \quad (7)$$

where F_{gg} is the driving force for grain growth. This parameter is associated with the grain growth and secondary recrystallization mechanisms, where the total amount of grain boundaries tends to be minimized. For example, if we consider grain growth in copper (boundary energy of $0.53 \text{ J}/\text{m}^2$) from $10 \mu\text{m}$ to $250 \mu\text{m}$, we obtain a value of $0.44 \text{ N}/\text{cm}^2$ for F_{gg} .

8.3. Driving force due to discontinuous precipitation

Discontinuous precipitation occurs when the high angle boundary migrates over a supersaturated matrix (α_0) and leaves behind it a mixture of phases ($\alpha+\beta$), frequently with lamellar morphology⁹⁵. It is fairly common that during annealing of strained supersaturated solid solutions recrystallization and discontinuous precipitation occur in a simultaneous and cooperative manner. The difference between the free energy due to supersaturation, ΔG_c , and the surface energy related to the interfaces of the precipitates, ΔG_o , give the equation:

Table 3. Driving forces for some phase transformations and solid state reactions⁹⁴.

Phase transformation	Driving force (J/mol)
Solidification of copper near to 0 K	- 13,000
Solid state precipitation at 1000 K	- 2,100
Homogenizing at 1000 K of Cu-40% Ni	- 1,900
Polymorphic transformation of tin (291 K)	- 2,200
Polymorphic transformation of iron (1183 K)	- 900
Recrystallization of work hardened copper	- 10.5 to - 50.6
Coalescence of fine particles	- 20
Grain boundary energy of material with average grain diameter of 30 μm and average grain boundary energy of 0.5 J/m ²	- 0.5

$$F_{\text{gg}} = (\Delta G_c / v) - (2f\sigma / \lambda) \quad (8)$$

Being that:

$$(\Delta G_c / v) \sim (2f\sigma / \lambda) \quad (9)$$

where v is the molar volume, f represents the volume fraction of precipitates, λ is the spacing between precipitates, and σ is the interfacial energy matrix/precipitate. For small solute concentrations c_o , the ΔG_c parameter can be written⁹⁵ as follows:

$$\Delta G_c = R \cdot (T - T_1) \cdot c_o \cdot \ln c_o \quad (10)$$

where T is the annealing temperature, T_1 is the temperature at which all solute is fully in solid solution, and R is the gas constant (8.314 J/mol.K). For example, if we consider that 4.9 at. % of silver ($C_o = 0.049$) is fully soluble in copper at 779 °C (T_o) and that this alloy presents discontinuous precipitations at 300 °C, Equation 10 gives $5.39 \times 10^4 \text{ N/cm}^2$ for ΔG_c . Despite of this value being only an estimate, we can observe that it is very much greater than the driving force for recrystallization.

8.4. Retarding forces due to particles

A dispersion of fine particles can retard boundary motion. The reason for this is that part of the available driving force for boundary motion must be used to bypass the particles⁹⁶. The particle dispersion can be thought to exert a pinning force or opposing pressure on the moving boundaries (see Figure 11).

The amount of free energy dissipated depends on the boundary-particle interaction mechanism. Ashby, Harper and Lewis⁹⁸ identified two possible mechanisms:

- i) The boundary may *enter* and pass through the particle;
- ii) The boundary will *bend around* the particle enveloping and bypassing it.

The first mechanism was the basis of Zener's early calculations in 1948⁹⁹ that resulted in:

$$F_p = (3 \gamma V_{vp} / 2r) \quad (11)$$

Notice that although F_p is often called pinning force it actually has units of pressure.

The second mechanism was proposed by Rios¹⁰⁰ and gives a pinning force twice that of Zener's:

$$F_p = (3 \gamma V_{vp} / r) \quad (12)$$

Rios also showed¹⁰⁰⁻¹⁰² that the pinning force could be written in terms of the interface area per unit volume of particles, S_{vp} :

$$F_p = \gamma S_{vp} \quad (13)$$

Supposing an alloy containing 1 vol.% precipitates ($V_{vp} = 0.01$) of 5 μm radius with a grain boundary energy of $\gamma = 0.53 \text{ J/m}^2$, we obtain

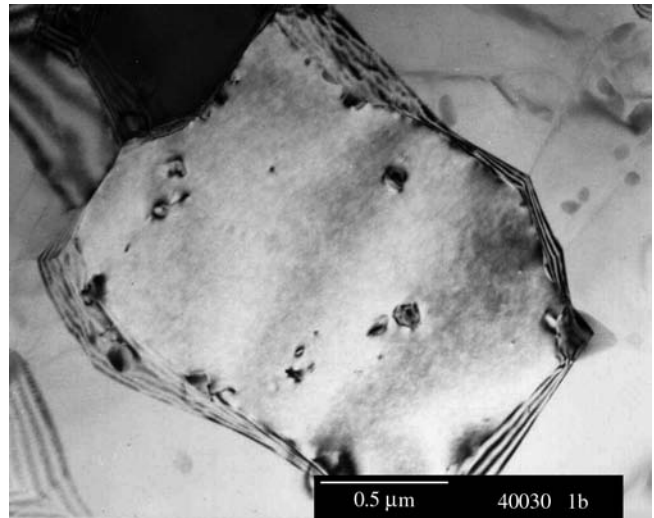


Figure 11. Transmission electron micrograph showing the interaction between intermetallic particles and migrating boundaries during recrystallization of an aluminum alloy⁹⁷.

with the help of Equation 11, the value of 0.16 N/cm² for Zener and 0.32 N/cm² for Rios. Much of the work concerning particle effects was carried out in the context of grain growth but the pinning force is the same regardless of the nature of the driving force moving the boundary.

8.5. Retarding force due to solute atoms

As a first (and coarse) approximation, we may extend the previous model of retarding force due to precipitates (F_p) to the retarding force due to solute atoms (F_s)⁹³. In this case, we use the atom fraction instead of volume fraction and the atomic radius instead of precipitate radii. If we take 1% of solute having an atomic radius of 0.125 nm (1.25 Å) in copper ($\gamma = 0.53 \text{ J/m}^2$), we obtain $F_p = 6.36 \times 10^3 \text{ N/cm}^2$. The extension of F_p (precipitates) to F_s (atoms) is strictly incorrect; for instance, particles do not move in the lattice, solute atoms can move by diffusion. This leads to two main consequences:

- i) There exists an interaction energy (U_b) between the boundary and the solute atom, which is very difficult to be measured or calculated. The solute concentration in the boundaries (C) increases relative to the average concentration (C_o): $C = C_o \cdot \exp(U_b/RT)$ ⁹³. This explains the large effect of small quantities of solutes on the recrystallization; and
- ii) Solute atoms tend to follow the moving grain boundaries by diffusion. In the case of very slow boundaries, these follow the boundary and exert little retarding force (this explains why

solute atoms have little effect on grain growth and a stronger effect on primary recrystallization). In the case of quick moving boundaries (very large driving forces) the boundary may break away from its impurities and move through the lattice almost unimpeded. The maximum retarding effect occurs for an intermediate range of grain boundary velocities^{38,93}.

8.6. Summation of forces acting during boundary migration

Starting from grain growth, the acting forces are F_{cg} , F_s e F_D , and the necessary condition for grain growth to occur is:

$$F_{gg} > F_s + F_p \quad (14)$$

On recrystallization, the forces acting at the reaction front are shown in Figure 12. The necessary condition for recrystallization to occur is that the summation of the driving forces is greater than the summation of the retarding forces, i.e.:

$$F_N + F_C > F_s + F_D \quad (15)$$

9. Experimental Evaluation of the Growth Velocity

Several microstructural analysis methods have been used to evaluate the growth velocity during recrystallization¹⁰⁴. Nowadays, the most accepted one is the method proposed by Cahn and Hagel¹⁰⁵, who demonstrated that the average migration velocity (V_{CH}) of the migrating reaction fronts, i.e., of the large angle grain boundaries between the recrystallized and non-recrystallized areas, can be obtained from quantitative stereological determinations performed on the polished metallographic plane, with the help of equation:

$$V_{CH} = (1 / S_{vr}) \cdot (dV_{vr} / dt) \quad (16)$$

where S_{vr} is the interfacial area per unit volume between recrystallized and non-recrystallized regions, V_{vr} is the recrystallized volume fraction, and t is the annealing time.

The analysis that has been performed to this point did not take into account two important characteristics of the recrystallization phenomena on the growth rate of the recrystallized regions. The first one is that the distribution of crystalline defects after plastic deformation is very heterogeneous, i.e., the stored energy of the crystalline defects, which is the main driving force for recrystallization, may vary considerably with position within the same grain and from one grain to another.

The second characteristic is that the driving force decreases with time during recrystallization, due to concurrent recovery in the non-recrystallized regions. This competition between recovery and recrystallization is more pronounced in metals and alloys with a BCC crystal structure and in the metals with a FCC crystal structure with high SFE. In these materials, the softening fraction due to recovery and recrystallization is substantially higher than the recrystallized volume fraction¹⁰⁶.

Both the strain heterogeneity and the occurrence of recovery may lead to a decrease in the speed of grain boundary migration during recrystallization (see Figure 13) and even to the recrystallization stagnation, both leading to deviations¹⁰⁷⁻¹¹⁰ in the KJMA equation.

DeHoff¹¹¹ gave an important idea to the analysis of recrystallization kinetics: the concept of the microstructural path. He proposed that the recrystallization follows a path in the S_v vs. V_v space. He

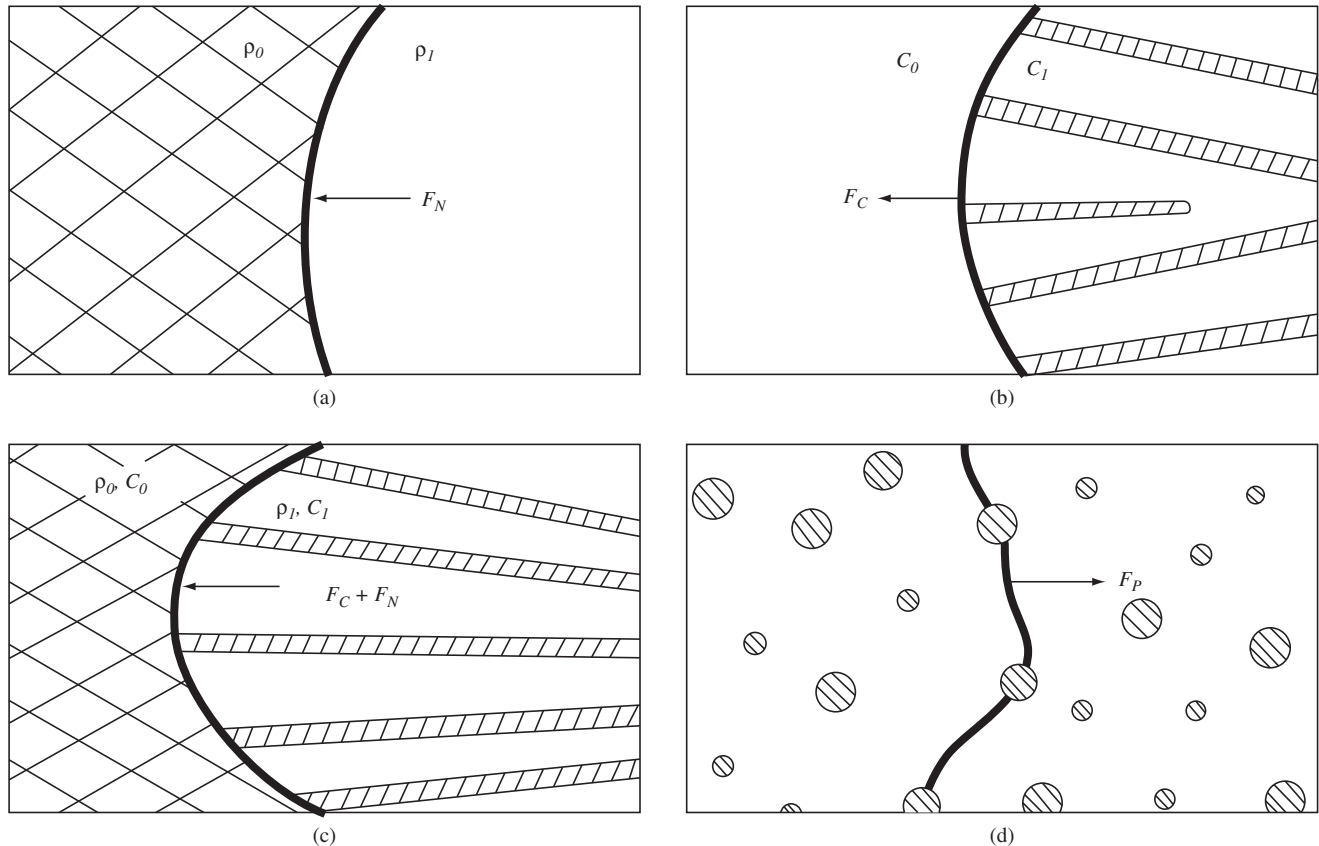


Figure 12. Forces acting at the reaction front. a) Recrystallization: grain boundary is the reaction front, reducing dislocation density from ρ_0 to ρ_1 ; b) Discontinuous precipitation: grain boundary is the reaction front, reducing supersaturation from C_0 to C_1 ; c) Discontinuous precipitation combined with recrystallization. Dislocation density ρ_0 and supersaturation C_0 reduced to ρ_1 and C_1 ; and d) Dispersed particles exerting retarding force on grain boundary migration^{53,103}.

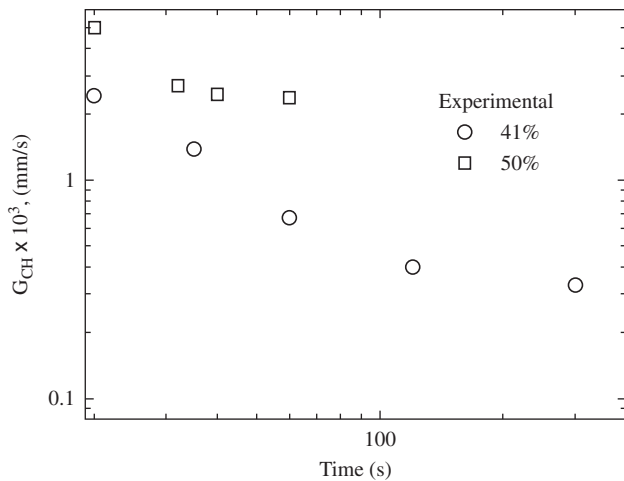


Figure 13. Variation of the average migration rate (V_{CH}) with annealing time at 400 °C for two levels of cold working (41 and 50%) in the AA3003 aluminum alloy^{113,114}.

demonstrated the usefulness of measuring S_v as a function of time in addition to V_v . The microstructural path method has been subsequently developed and extensively employed, notably by Vandermeer and co-workers^{104,112}.

As a final comment, it should be mentioned that the current development of 3-D metallography⁵¹ is making possible detailed *in situ* studies (and in relatively thick samples) of the growth in individually recrystallized regions.

10. Final Remarks

Some 120 years passed after Kalisher used for the first time in 1881 the technical term or keyword *recrystallization*, and we may say that a large volume of scientific and technological knowledge has been accumulated. Although various scientific aspects still need further clarification, the available knowledge already allows us a satisfactory control over the microstructure of metallic materials during its thermomechanical processing.

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